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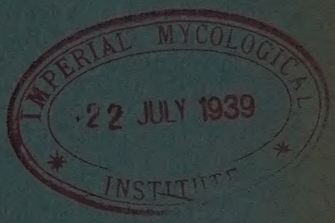

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TABLE DES MATIÈRES

Pages

MATHÉMATIQUES

D. Raikov. On the Composition of Analytic Distribution Functions . . 511

THÉORIE DE L'ÉLASTICITÉ

G. N. Savine. Tensions dans un plan élastique avec une série infinie de coupures égales 515

PHYSIQUE DU GLOBE

W. W. Shoulejkin, Corresponding Member of the Academy. Connexion between the Elements of the Monsoon Field and the Heat Balance of the Sea 519

W. W. Shoulejkin, Corresponding Member of the Academy. A More Precise Determination of Humidity from the Records of Aerological Sounds 525

PHYSIQUE CHIMIQUE

J. B. Chariton and **B. M. Stepanov.** On the Transition of the Combustion of Explosives into Detonation 527

J. B. Chariton and **E. V. Rdultovskaja.** On the Transmission of Detonation from Gases to Explosives 530

CHIMIE

Anna Gelman. Complex Compounds of Platinum and Butadiene (Divinyl) 532

O. E. Zviagincev and **V. G. Tronev.** Oxidation of CuS and Transference of Cu into Aqueous Solution at Air Pressure 537

V. G. Tronev and **S. M. Bondin.** Oxidation of ZnS and Transference of Zn into Aqueous or Alkali Solution at Air Pressure 541

V. I. Goremykin and **K. A. Gladyshevskaja.** On Mixed Platinum Hydroxylamine Tetramines 544

P. D. Dankov. Regularities of Formation and Structure of Protective Films on Metals 548

GÉOLOGIE

G. G. Kazakashvili and **P. A. Topuria.** On Some Syenite Rocks of the Supsa River (West Georgia) 553

PÉTROGRAPHIE

B. M. Kupletsky and **O. I. Kovaleva.** Essay of a Rational Quantitative Mineralogical Classification of Granitoids 557

CRISTALLOCHIMIE

V. A. Frank-Kameneckij. The Course of Orientated Displacement of Magnetite by Iron 561

SCIENCE DU SOL

I. D. Sedletzky. Gedroizite in the Alkali-Soils 565

GÉNÉTIQUE

E. P. Panfilova. Difference in Blood Value of Karakul Sheep in Correlation with Their Organic Constitution 569
M. D. Rcheulishvili. Variation of the Shape of the «Kurdjuk» and of the Tail Appendix in Tushin Sheep 573
M. D. Rcheulishvili. Variation in Tail Form in Imeritian Sheep . . . 576

PHYSIOLOGIE VÉGÉTALE

I. A. Filippenko. Yarovization of Seed of Agricultural Plants by Immersion in Water 580

ZOOLOGIE

A. N. Svetovidov. Contribution to the Comparative Anatomy of the Genus *Muraenolepis* (*Gadiformes*) 583
Z. M. Ptchelina. *Relictogobius Kryžanovskii* n. g. n. sp., un nouveau gobiidé provenant d'un lac salé de la presqu'île Abraou (bassin de la Mer Noire) 586

MORPHOLOGIE EXPÉRIMENTALE

B. G. Novikov. Experimental Analysis of Development of Plumage Coloration in *Perdix perdix* L. 590

PHYSIOLOGIE DES ANIMAUX

I. Beritov, Member of the Academy. On the Physiological Properties of the Nerveless Region of the Skeletal Muscle 592
I. Beritov, Member of the Academy. On the Origin of the Acetylcholine Contracture 597

EMBRYOLOGIE DES ANIMAUX

T. I. Privolnev and **A. M. Rasumovsky.** Influence of Low Temperature on Different Stages of Development of Eggs of Bream (*Abramis brama*) 602

PALÉOZOOLOGIE

V. I. Zubareva. A New Form of a Bird from the Pliocene of Odessa . 606
I. G. Pidoplichka. Contribution to the History of the Fauna of the USSR 609

Rédacteur en chef A. Fersman. Rédacteur S. Bernstein.

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MATHEMATICS

ON THE COMPOSITION OF ANALYTIC DISTRIBUTION FUNCTIONS

By D. RAIKOV

(Communicated by A. N. Kolmogoroff, Member of the Academy, 28. III. 1939)

The present note contains a result pertaining to the problem of the dependence of the analytical character of the composition of distribution functions on the analytical character of its components*. It is known that the composition of a continuous distribution function with an arbitrary distribution function is itself a continuous distribution function, and that if one of the components is absolutely continuous, the composition is absolutely continuous, too. It is, moreover, known that the maximum of the derivative of the composition cannot exceed the maxima of the derivatives of its components. On ground of these and similar facts Paul Lévy has formulated the following principle (1): «Toute condition de continuité ou d'analycité imposée à une seule des lois composantes, toute condition limitant l'irrégularité de sa fonction de répartition et des dérivées de cette fonction, est vérifiée pour la loi résultante. La composition ne peut qu'améliorer la continuité et la régularité».

From the result proved below it will, however, follow that this principle does not hold in so general a form. We shall namely show that there exist two integral analytic distribution functions whose composition is not analytic at the origin.

We shall consider distribution functions $F(x)$ regular in some strip $|Ix| < R$. If $F(x)$ is bound in every interior strip $|Ix| \leqslant r < R$, then its composition with any distribution function $G(x)$ will be regular in the strip $|Ix| < R$ and bound in every interior strip. In fact, in virtue of the condition the expression $\int_{-\infty}^{\infty} F(x-y)\,dG(y)$ has sense for all x situated on any straight line $Ix = \pm r$, $|r| < R$, and it may be easily shown that the function defined by this expression possesses the asserted properties.

* Distribution functions or laws are non-decreasing functions $F(x)$ defined for all x, $-\infty < x < \infty$, and subjected to the conditions $F(x) \to 0$ for $x \to -\infty$ and $F(x) \to 1$ for $x \to +\infty$. The composition (Faltung) $F(x) = F_1(x) \ast F_2(x)$ of two distribution functions $F_1(x)$ and $F_2(x)$ is defined as the distribution function

$$F(x) = \int_{-\infty}^{\infty} F_1(x-y)\,dF_2(y) = \int_{-\infty}^{\infty} F_2(x-y)\,dF_1(y).$$

$F_1(x)$ and $F_2(x)$ are called its components.

But there exist analytic distribution functions not bound in any strip however narrow $|Ix| < \rho$. Such is, for instance, the integral distribution function *

$$F(x) = e^{1-e^{e^{-x}}}.$$

In fact, putting $x = \xi + i\eta$, we have

$$|F(x)| = e^{1-e^{e^{-\xi}\cos\eta}\cos(e^{-\xi}\sin\eta)},$$

and for $\xi \to -\infty$ and $|\eta| < \frac{\pi}{2}$ this expression will not be bound, since $e^{e^{-\xi}\cos\eta} \to \infty$ and $\cos(e^{-\xi}\sin\eta)$ oscillates between $+1$ and -1.

Theorem. *If $F(x)$ is a distribution function which is not analytic and bound in any strip $|Ix| < \rho$, then the «symmetrized» distribution function*

$$F^*(x) = F(x) \ast \overline{F}(x) = F(x) \ast [1 - F(-x)]$$

is not regular at $x = 0$.

The proof is based on two lemmas on distribution functions with real non-negative characteristic functions **.

Lemma 1. *Let $F(x)$ be a distribution function and $f(t)$ its characteristic function. If $F'(0)$ exists and $f(t) \geqslant 0$, then the integral $\int_{-\infty}^{\infty} f(t)\,dt$ converges and $F'(x)$ exists for all real x and*

$$F'(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-ixt} f(t)\,dt.$$

Lemma 2. *Let $F(x)$ be a distribution function and $f(t)$ its characteristic function. If $F(x)$ is regular in $|x| < R$ and $f(t) \geqslant 0$, then the integral $\int_{-\infty}^{\infty} f(t)\,e^{r|t|}\,dt$ converges for all $r < R$ and $F(x)$ is regular in the strip $|Ix| < R$ and bound in any interior strip $|Ix| \leqslant r < R$.*

Proof of Lemma 1. To the distribution function

$$F_h(x) = \frac{1}{2h}\int_{-h}^{h} F(x+\tau)\,d\tau$$

corresponds the characteristic function $f(t)\frac{\sin ht}{ht}$. By the inversion formula of P. Lévy

$$F_h(x) - F_h(-x) = \lim_{\omega\to\infty} \frac{1}{2\pi}\int_{-\omega}^{\omega} f(t)\,\frac{\sin ht}{ht}\,\frac{2\sin xt}{t}\,dt,$$

whence, putting $x = h$, we obtain

$$\frac{F_h(h) - F_h(-h)}{2h} = \frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)\left(\frac{\sin ht}{ht}\right)^2 dt. \qquad (1)$$

---

* This example has been pointed out by A. Gelfond.

** The characteristic function of a distribution function is defined as

$$f(t) = \int_{-\infty}^{\infty} e^{itx}\,dF(x), \quad -\infty < t < \infty.$$

But

$$\frac{F_h(h)-F_h(-h)}{2h}=$$

$$=\frac{1}{2h}\int_0^h\left(\frac{h+\tau}{h}\,\frac{F(h+\tau)-F(-h-\tau)}{2(h+\tau)}+\frac{h-\tau}{h}\,\frac{F(h-\tau)-F(-h+\tau)}{2(h-\tau)}\right)d\tau=$$

$$=F'(0)+o(h)$$

for $h\to 0$. Thus, the limit of the right hand side of (1) for $h\to 0$ exists, and since $f(t)$ is real non-negative it must be equal to

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)\,dt.$$

The last assertion of the Lemma is then obtained by differentiating the inversion formula

$$F(x)-F(0)=\frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)\,\frac{1-e^{-ixt}}{it}\,dt.$$

Proof of Lemma 2. Since $F'(0)$ exists, we have, by Lemma 1,

$$\int_{-\infty}^{\infty} f(t)\,dt<\infty$$

and

$$F'(x)=\frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)\,e^{-ixt}\,dt, \tag{2}$$

i. e., since $f(t)$ is real non-negative, that $F'(x)$ coincides up to a constant positive factor with the characteristic function of a certain distribution law. Since, on the other hand, $F'(x)$ is regular in $|x|<R$, the representation (2) for $F'(x)$ holds in the whole strip $|Ix|<R$ (2), whence follows the first assertion of the Lemma. Further, putting $x=\xi+i\eta$, $|\eta|<R$, we have

$$F(\xi+i\eta)=F(\xi)+\int_{\xi}^{\xi+i\eta} F'(x)\,dx,$$

and thus, for $|\eta|\leqslant r(<R)$,

$$|F(x)|\leqslant 1+\frac{r}{2\pi}\int_{-\infty}^{\infty} f(t)\,e^{r|t|}\,dt,$$

which proves the second assertion of the lemma.

Proof of the Theorem. It is easily seen that if for a certain distribution function $F(x)$ the symmetrized distribution function $F^*(x)=F(x)*\overline{F}(x)$ is regular in $|x|<R$, then $F(x)$ is regular in the strip $|Ix|<\frac{R}{2}$ and bound in every interior strip $|Ix|\leqslant\rho<\frac{R}{2}$. In fact, the characteristic function of $F^*(x)$ is $|f(t)|^2\geqslant 0$ and, by Lemma 2,

$$\int_{-\infty}^{\infty}|f(t)|^2\,e^{r|t|}\,dt<\infty$$

for all $r < R$. Let r' be a number between r and R. By Schwarz inequality we have

$$\left(\int_{-\infty}^{\infty} |f(t)|\, e^{\frac{r}{2}|t|}\, dt\right)^2 = \left(\int_{-\infty}^{\infty} |f(t)|\, e^{\frac{r'}{2}|t|}\, e^{-\frac{r'-r}{2}|t|}\, dt\right)^2 \leqslant$$

$$\leqslant \int_{-\infty}^{\infty} |f(t)|^2 e^{r'|t|}\, dt \int_{-\infty}^{\infty} e^{-(r'-r)|t|}\, dt < \infty.$$

Thus, the integral

$$\int_{-\infty}^{\infty} |f(t)|\, e^{\rho|t|}\, dt$$

converges for all $\rho < \frac{R}{2}$. Hence it follows that $F'(x)$ defined by (2) is regular in the strip $|Ix| < \frac{R}{2}$ and bound in every interior strip $|Ix| \leqslant \rho < \frac{R}{2}$. But then, similar as in the proof of the second part of Lemma 2, we find that the same is true for $F(x)$.

Our theorem is thus proved, since the distribution functions considered in it are not bound in any strip $|Ix| < \rho$.

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THÉORIE DE L'ÉLASTICITÉ

TENSIONS DANS UN PLAN ÉLASTIQUE AVEC UNE SÉRIE INFINIE DE COUPURES ÉGALES

Par G. N. SAVINE

(Présenté par S. L. Soboleff, de l'Académie, le 23.III.1939)

§ 1. Supposons que le milieu élastique remplit le plan oxy, possédant une série illimitée de coupures congrues d'une forme arbitraire, situées de manière que chaque coupure suivante s'obtient de la précédente par la translation de 2π le long de l'axe ox. Au contour de chaque coupure sont appliquées les forces, égales dans les points correspondants $\overline{X}_n$ $\overline{Y}_n$, et équivalentes sur chaque contour, au point de vue statique, à zéro.

Prenons comme origine de coordonnées un point situé à l'intérieur d'un des contours et divisons le plan oxy par les droites... $y=-3\pi$, $y=-\pi$; $y=-\pi$, $y=\pi$; $y=\pi$; $y=3\pi$, etc., en un nombre infini de bandes dont la largeur soit égale à 2π. Nous supposerons de plus, que chaque coupure est entièrement située à l'intérieur de la bande correspondante. Les composantes des tensions et des déformations seront les fonctions périodiques ayant la période 2π. Il suffit pour cela ne les considérer que dans la bande $-\pi \leqslant Rez \leqslant \pi$ avec la coupure donnée.

En appliquant la méthode habituelle [1], on peut démontrer que le problème posé n'a qu'une solution, à condition que 1) les composantes des tensions $\overline{X}_x$, $\overline{Y}_y$, $\overline{X}_y$ tendent uniformément vers zéro pour $y \to \infty$, et que 2) les composantes des déformations u, v soient des grandeurs bornées.

Ainsi, la résolution du problème posé s'est réduite à la détermination des composantes des tensions et des déformations dans la bande avec la coupure donnée, suivant le contour l de laquelle sont appliquées les forces extérieures données $\overline{X}_n$ et $\overline{Y}_n$.

Désignons par S la région limitée par les droites $y=-\pi$; $y=\pi$ et par le contour l de la coupure, de même que par S^* — la région à l'intérieur de l.

§ 2. On sait que la résolution du problème plan de la théorie de l'élasticité, d'après les forces extérieures $\overline{X}_n$ et $\overline{Y}_n$ données sur le contour, se réduit à la définition de deux fonctions analytiques de la variable complexe $\varphi(z)$ et $\psi(z)$, satisfaisant à la condition du contour

$$\overline{\varphi(z)}+\bar{z}\varphi'(z)+\psi(z)=-i\int_0^s(\overline{X}_n-i\overline{Y}_n)\,ds+C=f_1-if_2, \qquad (1)$$

où C désigne une constante complexe arbitraire. Les composantes des tensions $\overline{X}_x$, $\overline{Y}_y$ et $\overline{X}_y$ et des déformations u et v, les fonctions $\varphi(z)$ et

$\psi(z)$ étant trouvées, sont données par la formule:

$$\left.\begin{aligned} \overline{X}_x + \overline{Y}_y &= 4Re\{\varphi'(z)\}, \\ \overline{Y}_y - \overline{X}_x + 2i\overline{X}_y &= 2[\bar{z}\varphi''(z) + \psi'(z)], \end{aligned}\right\} \tag{2}$$

$$2\mu(u+iv) = \varkappa\varphi(z) - z\overline{\varphi'(z)} - \overline{\psi(z)}. \tag{3}$$

Les membres de gauche des équations (2) et (3) étant, comme il suit de la façon dont on a posé le problème, des fonctions périodiques de la période 2π, les fonctions $\varphi(z)$ et $\psi(z)$ doivent nécessairement être de la forme

$$\varphi(z) = \varphi_0(z);\ \psi(z) = \psi_0(z) - z\varphi_0'(z), \tag{4}$$

où $\varphi_0(z)$ et $\psi_0(z)$ sont les fonctions périodiques de la période 2π.

La condition limite (1) et les formules (2) et (3), donnant les tensions et les déformations comme fonctions de $\varphi_0(z)$ et $\psi_0(z)$, seront de la forme:

$$\overline{\varphi_0(z)} + (\bar{z} - z)\varphi_0'(z) + \psi_0(z) = f_1 - if_2, \tag{5}$$

$$\left.\begin{aligned} \overline{X}_x + \overline{Y}_y &= 4Re\{\varphi_0'(z)\}, \\ \overline{Y}_y - \overline{X}_x + 2i\overline{X}_y &= 2[(z-\bar{z})\varphi_0''(z) + \psi_0'(z) - \varphi_0'(z)] \end{aligned}\right\} \tag{6}$$

$$2\mu(u+iv) = \varkappa\varphi_0(z) - (z-\bar{z})\overline{\varphi_0'(z)} - \overline{\psi_0(z)}. \tag{7}$$

On peut prouver en partant du théorème de l'unicité et de la périodicité des fonctions $\varphi_0(z)$ et $\psi_0(z)$, que les fonctions $\varphi_0(z)$ et $\psi_0(z)$ sont déterminées à une constante complexe arbitraire C_1^* près:

$$\varphi_0(z) = C_1^*;\ \psi_0(z) = -\overline{C}_1^*. \tag{8}$$

Représentons la bande $-\pi \leqslant Rez \leqslant \pi$ sur le plan coupé suivant la partie négative de l'axe réel ox, au moyen de la fonction

$$w = e^{iz}. \tag{9}$$

Le contour de la coupure se transforme en un certain contour fermé L, ne contenant pas à l'intérieur l'origine des coordonnées. Désignons par $\sum$ la région infinie située hors du contour L, et par $\sum^*$ — la région située à l'intérieur de L.

Les fonctions $F_1(w) = \varphi_0(-i\ln w)$ et $F_2(w) = \psi_0(-i\ln w)$ sont holomorphes dans la région $\sum$ et remplient sur le contour L les conditions:

$$\overline{F_1(w)} - w(\overline{\ln w} + \ln w)F_1'(w) + F_2(w) = f_1 - if_2. \tag{10}$$

Déterminons les fonctions $F_1(w)$ et $F_2(w)$ par la méthode due à Mouskhelichvili (2). Multiplions les deux membres de l'équation (10) par $\frac{1}{2\pi i}\cdot\frac{dw}{w-w_e}$, où w_e est un point de la région $\sum^*$, et intégrons suivant le contour L dans le sens positif relativement à la région $\sum$. On trouve

$$\begin{aligned} &\frac{1}{2\pi i}\int_L \frac{\overline{F_1(w)}}{w-w_e}dw - \frac{1}{2\pi i}\int_L \frac{w(\overline{\ln w} + \ln w)}{w-w_e}F_1'(w)\,dw = \\ &= \frac{1}{2\pi i}\int_L \frac{f_1 - if_2}{w-w_e}dw = A_1 - iA_2 + \overline{C}. \end{aligned} \tag{11}$$

En faisant dans l'expression (11) $w_e \to w_0$, où w_0 est un point quelconque sur le contour L, et en ajoutant à (11) les égalités évidentes

$$\left.\begin{aligned} -\frac{1}{2}\overline{F_1(w)} - \frac{1}{2\pi i}\int_L \frac{\overline{F_1(w)}}{\overline{w}-\overline{w}_e}d\overline{w} &= 0, \\ -\frac{w_0(\overline{\ln w_0} + \ln w_0)}{2}F_1'(w_0) + \frac{w_0(\overline{\ln w_0} + \ln w_0)}{2\pi i}\int_L \frac{F_1'(w)}{w-w_e}dw &= 0, \end{aligned}\right\} \tag{12}$$

nous trouvons, après les transformations faciles:

$$\overline{F_1(w_0)} - \frac{1}{2\pi i}\int_L \overline{F_1(w)}\, d\left\{\ln\frac{w-w_0}{\overline{w}-\overline{w}_0}\right\} + \\ + \frac{1}{2\pi i}\int_L F_1(w)\, d\left\{\frac{w_0 \ln(\overline{w}_0 w_0) - w\ln(\overline{w}w)}{w-w_0}\right\} = -A_1^{(e)} + iA_2^{(e)} - \overline{C}, \tag{13}$$

où $A_1^{(e)} - iA_2^{(e)} + \overline{C}$ est la limite du membre de droite de l'équation (11), lorsque w_e tend de l'intérieur du contour vers le point w_0 du contour L.

L'équation (13) est une équation intégrale de Fredholm de 2-me catégorie au noyau régulier. L'équation (13) n'est pas résolvable, car l'équation homogène (13) a une solution non triviale. Transformons d'après Chermann [3] notre équation (13) en ajoutant au membre de gauche l'opérateur

$$i\overline{\ln w}\, Re\left\{\frac{b}{2\pi}\int_L \frac{F_1(w)\,dw}{(w-b)^2}\right\}, \tag{14}$$

où b est un certain point à l'intérieur $\sum^*$.

L'équation donnant la solution de notre problème aura donc la forme:

$$\overline{F_1(w)} - \frac{1}{2\pi i}\int_L \overline{F_1(w)}\, d\left\{\ln\frac{w-w_0}{\overline{w}-\overline{w}_0}\right\} + \\ + \frac{1}{2\pi i}\int_L F_1(w)\, d\left\{\frac{w_0\ln(\overline{w}_0 w_0) - w\ln(\overline{w}w)}{w-w_0}\right\} + \\ + i\overline{\ln w}\, Re\left\{\frac{b}{2\pi}\int_L \frac{F_1(w)}{(w-b)^2}\,dw\right\} = -A_1^{(e)} + iA_2^{(e)} - \overline{C}_1. \tag{15}$$

§ 3. L'étude de l'équation (15) peut être effectuée comme l'a fait Mouskhelichvili [4]. Supposons que l'équation (15) admet une solution. En introduisant dans les fonctions considérées:

$$\Phi(w_e) = \frac{1}{2\pi i}\int_L \frac{F_1(w)}{w-w_e}\,dw \tag{16}$$

et

$$\Psi(w_e) = -\frac{1}{2\pi i}\int_L \frac{\overline{F_1(w)}}{w-w_e}\,dw + \frac{1}{2\pi i}\int_L \frac{w\ln(\overline{w}w)}{w-w_e}F_1'(w)\,dw + A_1 - iA_2 + \overline{C}, \tag{17}$$

nous aurons l'équation

$$\overline{\Phi(w)} + w_0(\overline{\ln w}_0 - \ln w_0)\Phi'(w) - \Psi(w) + i\overline{\ln w}\, Re\left\{\frac{b}{2\pi}\int_L \frac{F_1(w)}{(w-b)^2}dw\right\} = 0. \tag{18}$$

Les fonctions $\Phi(w)$ et $\Psi(w)$ sont, comme il suit de (16) et de (17), des fonctions holomorphes dans la région $\sum^*$. L'équation (18) à la variable z est de la forme

$$\overline{F(z)} + (z-\overline{z})F'(z) - G(z) + \overline{z}\, Re\{F'(a)\} = 0, \tag{19}$$

où $\Phi(w) = F(z)$; $\Psi(w) = G(z)$ et $a = -i\ln b$.

En multipliant les deux membres de l'équation (19) par dz et en intégrant suivant le contour l, nous obtiendrons

$$Re\{F'(a)\} = 0, \tag{20}$$

d'où l'on a

$$\overline{F(z)} + (z-\overline{z})F'(z) - G(z) = 0. \tag{21}$$

En désignant $F(z) = iF^*(z)$; $G(z) - zF'(z) = iH^*$, nous pouvons écrire l'équation (21) de la manière suivante:

$$\overline{F^*(z)} + \overline{z}F^{*\prime}(z) + H^*(z) = 0. \tag{22}$$

L'équation (22) correspond au problème limite de la théorie de l'élasticité avec des forces extérieures, appliquées sur le contour, égales à zéro. D'après le théorème de l'unicité (1), les fonctions $F^*(z)$ et $H^*(z)$ sont de la forme

$$F^*(z) = iC_1 z + C_2;\ H^*(z) = -\overline{C_2} \tag{23}$$

où C_1 est une constante réelle et C_2 — une constante complexe. D'où, par conséquent

$$F(z) = -C_1 z + iC_2;\ G(z) = -C_1 z - i\overline{C_2}. \tag{24}$$

Il suit des expressions (20) et (24) que $C_1 \equiv 0$, c'est-à-dire

$$F(z) = iC_2;\ G(z) = -i\overline{C_2}. \tag{25}$$

On a d'après (16), (17) et (25):

$$\left.\begin{aligned} \Phi(w_e) &= \frac{1}{2\pi i}\int_L \frac{F_1(w)}{w - w_e}\,dw = iC_2, \\ \Psi(w_e) &= -\frac{1}{2\pi i}\int_L \frac{\overline{F_1(w)}}{w - w_e}\,dw + \frac{1}{2\pi i}\int_L \frac{w \ln (\overline{w} w)}{w - w_e} F_1'(w)\,dw + \\ &\quad + A_1 - iA_2 + \overline{C} = -i\overline{C_2}. \end{aligned}\right\} \tag{26}$$

En introduisant une nouvelle fonction $F_1^* = F_1 + iC_2$ et en la substituant dans l'équation (26), nous trouvons que la fonction F_1^* est holomorphe dans la région $\sum$ et satisfait à l'équation (11), dans laquelle au membre de droite est ajoutée une constante $2i\overline{C_2}$, c'est-à-dire

$$\left.\begin{aligned} &\frac{1}{2\pi i}\int_L \frac{F_1^*(w)}{w - w_e}\,dw = 0, \\ &\frac{1}{2\pi i}\int_L \frac{\overline{F_1^*(w)}}{w - w_e}\,dw - \frac{1}{2\pi i}\int_L \frac{w \ln (\overline{w} w)}{w - w_e} F_1^{*\prime}(w)\,dw = \\ &\quad = A_1 - iA_2 + \overline{C} + 2i\overline{C_2}. \end{aligned}\right\} \tag{27}$$

Notons que la constante C_2 disparaît lorsque nous transformons $F_1^*(w)$ par une intégrale de Cauchy dans la région $\sum$. Par conséquent, la résolution du problème n'exige pas la détermination de cette constante.

On peut prouver que le problème considéré n'a qu'une seule solution, qui peut être obtenue de l'équation intégrale (15) qui n'admet qu'une solution. Cette affirmation peut être démontrée d'après Chermann (5).

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GEOPHYSICS

CONNEXION BETWEEN THE ELEMENTS OF THE MONSOON FIELD AND THE HEAT BALANCE OF THE SEA

By W. W. SHOULEJKIN, Corresponding Member of the Academy

In the preceding works ([1], [2], [3]) we have found expressions for the various elements of the monsoon field on the ground of a hypothetic law of variation of the radial wind components with the growth of distance from the centre of a circular sea (or a circular continent). At the end of the deduction the good accordance of the results of the original assumptions with reality was verified. The coincidence proved to be highly satisfactory. But in the theory suggested one substantial moment was still lacking, the elements of the monsoon field had not been connected with the heat balance of the sea; while just the heat balance of the latter as of the accumulator of heat determines all the system of monsoon phenomena under natural conditions.

In the present work we shall try and fill up this gap and to determine the connexion sought for.

Let us imagine that the globe is deprived of oceans and that the atmosphere around it has assumed a certain temperature stabilized by the existing heat radiation of the sun. Let us consider this temperature as zero in the further calculations. Let us suppose that one single sea appeared on such a solid globe. The winter temperature consequently increased by τ, the «anomaly» τ naturally being different for different points above the sea and the continent. Now we shall examine the heat processes which will arise in the atmosphere under the influence of the sea.

It proved previously([4]) possible to describe the process of transferring of heat by the monsoon currents as a certain statistic process of turbulent conduction of heat, and the quantitative results obtained in this way for the heat currents from the sea to the continent coincided with the results of the calculations of the same currents on the ground of the hydrodynamic picture ([1]).

Now let us replace the atmosphere by its «heat model», i. e. a thin film having a certain conventional conductivity AH([4]). Then we shall be able to say that each cm^2 of the underlying surface gives away (or receives, depending on the sign of the derivatives) an amount of heat equal to

$$AH \cdot \nabla^2 \tau.$$

Taking the same unit of area, our two-dimensional model of the atmosphere will lose a certain amount of heat $\sigma\tau$ in the extra-heat radiation (besides the loss in the case of a purely solid globe, the temperature of the atmosphere above which has been considered as zero). Taking into account

the small conductivity and heat capacity of the rocks covering the continents, we shall need only the two mentioned terms for the calculation of the heat balance of the atmosphere above the continent. Above the sea we shall have to consider one more term—the amount of heat given to the atmosphere by every cm² of the water surface. Generally speaking, we may take it equal to

$$\alpha\ (\tau_w - \tau),$$

where τ_w denotes the temperature of the water surface which will be considered constant for all the surface of the sea. This expression is the nearer to truth, the more the heat given away by the sea in the heat interchange with the atmosphere prevails in comparison with the other elements of the heat balance.

Consequently, for the atmosphere above the sea we shall have

$$(\nabla^2 - \varkappa_1^2)\,\tau + b^2 = 0 \qquad (1)$$

and for the atmosphere above the land

$$(\nabla^2 - \varkappa_2^2)\,\tau = 0, \qquad (2)$$

using the following abbreviations

$$\varkappa_1^2 = \frac{\sigma+\alpha}{AH}; \quad \varkappa_2^2 = \frac{\sigma}{AH}; \quad b^2 = \frac{\alpha\tau_w}{AH}. \qquad (3)$$

In the present article we shall examine only the case of a circular sea (and a circular island). The radius of the sea will be R; in its centre let us place the pole of the system of co-ordinates. Then the symbolic equations (1) and (2) will be written in their full form

$$\frac{d^2\tau_1}{dr^2} + \frac{1}{r}\frac{d\tau_1}{dr} - \varkappa_1^2\tau_1 + b^2 = 0 \qquad (4)$$

$$\frac{d^2\tau_2}{dr^2} + \frac{1}{r}\frac{d\tau_2}{dr} - \varkappa_2^2\tau_2 = 0. \qquad (5)$$

For better clearness τ_1 denotes the heat anomalies within the circle with the radius R and τ_2 those without it. The limit conditions of the problem obviously are

$$\text{with } r = R \begin{cases} \tau_1 = \tau_2 \\ \dfrac{d\tau_1}{dr} = \dfrac{d\tau_2}{dr} \end{cases} \qquad (6)$$

Under these limit conditions the integrals of equations (4), (5) will be written in the very simple form

$$\frac{\tau_1}{\Theta} = 1 - N_1 I_0(\varkappa_1 r), \qquad (7)$$

$$\frac{\tau_2}{\Theta} = N_2 K_0(\varkappa_2 r). \qquad (8)$$

Here as usually in the Bessel and Macdonald functions of imaginary argument the following denominations are used:

$$I_n(x) = i^{-n} J_n(ix)$$

$$K_n(x) = \frac{\pi}{2}\ \lim\{[I_{-n}(x) - I_n(x)]\ \operatorname{cosec}\ \pi n\}_{n \to \text{entir}}$$

The constants N_1 and N_2 are expressed as the functions of the radius R of the sea:

$$N_1 = \frac{\mu K_1(\varkappa_2 R)}{(I_1(\varkappa_1 R)\,K_0(\varkappa_2 R) + \mu I_0(\varkappa_1 R)\,K_1(\varkappa_2 R)}, \qquad (9)$$

$$N_2 = \frac{I_1(\varkappa_1 R)}{I_1(\varkappa_1 R)\,K_0(\varkappa_2 R) + \mu I_0(\varkappa_1 R)\,K_1(\varkappa_2 R)}. \qquad (10)$$

The value

$$\Theta = \frac{b^2}{x_1^2} = \frac{\tau_w}{1 + \frac{\sigma}{\alpha}} \qquad (11)$$

as it is easy to see, has a curious physical meaning: it is the temperature that would have been assumed by the «model» of the atmosphere if all the globe had been covered with ocean.

The coefficient μ is connected with the constant of radiation σ and that of the heat interchange α:

$$\mu = \frac{x_2}{x_1} = \sqrt{\frac{\sigma}{\sigma + \alpha}}. \qquad (12)$$

The constant σ can be determined now with sufficient accuracy on the base of the formulae (12), (13), (23) and (22) of (4), verified by means of the diagram in Fig. 19 of the cited paper. The constant α is also known approximately on the base of many measurements of the thermal interchange between the sea and the atmosphere (5). Namely:

$$\left.\begin{aligned} \sigma &= 6.8 \cdot 10^{-5} \\ AH &= 4 \cdot 10^{12} \\ \alpha &= 3.3 \cdot 10^{-4} \end{aligned}\right\} \qquad (13)$$

Therefore

$$\mu = 0.41$$

Applying the relations (7), (8), (9), (10), it is easy to determine the connexion between the radius of the sea and the anomalies of the air temperature caused by the sea: in the centre of the sea and on the coast. Indeed, substituting in (7) $r = 0$ we shall find for the temperature anomaly over the centre of the sea:

$$\frac{\tau_0}{\Theta} = 1 - N_1. \qquad (14)$$

Then taking in (8) or (7) $r = R$, we shall have for the anomaly over the coast:

$$\frac{\tau_R}{\Theta} = N_2 K_0(x_2 R). \qquad (15)$$

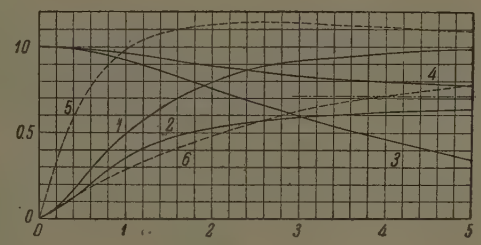

Fig. 1.

Placing here instead of N_1 and N_2 their expressions (9), (10) and having made the calculations according to the tables of the functions I and K, we shall get the dependences sought for which are represented in Fig. 1. The curve *1* corresponds here to the temperature over the centre of the sea, and the curve *2* to that over the coast. The temperature anomaly τ is expressed in the parts of the maximum anomaly Θ (axis of the ordinates). The radius of the sea is expressed in thousand kilometers (abscissa-axis).

Now let us examine the case of a circular island, the last remainder of land on a globe covered with water. Let the radius of the island be R and the pole of the system of co-ordinates be in its centre. Let us keep the index «1» for the values corresponding to the zone $0 < r < R$ and «2» — for the external zone. Then the term b^2 will be transferred

from equations (1) and (4) respectively to equations (2) and (5). Their integrals (under the same limit conditions) will be

$$\frac{\tau_1}{\Theta} = N_1 I_0(x_1 r); \tag{16}$$

$$\frac{\tau_2}{\Theta} = 1 - N_2 K_0(x_2 r). \tag{17}$$

The expressions of the constants N_1 and N_2 will remain the same (9), (10), but we shall have to keep in mind that now

$$x_1^2 = \frac{\sigma}{AH}; \quad x_2^2 = \frac{\sigma + \alpha}{AH} \tag{18}$$

and therefore

$$\mu = \sqrt{\frac{\sigma + \alpha}{\sigma}}. \tag{19}$$

Remembering the values σ and α from (13), we get in the case of the island:

$$\mu = 2.43. \tag{19'}$$

On the diagram Fig. 1 are plotted the curves calculated by means of (16), (17) and (20) for the temperature anomalies: above the centre of the island (curve *3*) and above the shore of the island (curve *4*). The dimensions of the island are expressed in thousand kilometers. It is easy to show that with unlimited increase of the dimensions of the sea or the island, the temperature anomaly above the shore tends to a common limit (marked in the Fig. 1 with a straight line):

$$\lim \left| \frac{\tau_R}{\Theta} \right|_{R \to \infty} = \frac{1}{1 + \sqrt{\frac{\sigma}{\alpha + \sigma}}}. \tag{20}$$

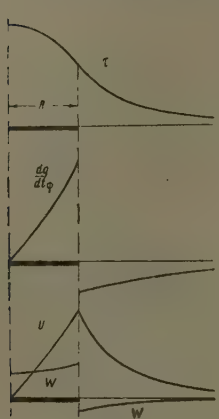

Fig. 2.

Fig. 2 represents the curve τ of the distribution of the temperature anomalies above the sea and the surrounding continent. This curve has been calculated according to formulae (7)—(10) in application to the case $x_1 R = 1$.

It is easy to prove that it has a close resemblance in form to the corresponding curve calculated in work [1] by means of an utterly different method. Only the point of inflexion in the new version naturally fell just over the coast line. The sea is denoted as previously by a thick black stripe under the axis of the abcissae. In order to get the distribution of elements in space, one has to imagine the whole picture revolving about the axis of ordinates.

Having learnt the distribution of τ, is is easy to calculate the amount of heat $\left(\frac{dq}{dt}\right)_\psi$ taken by the atmosphere from a section of the sea surface, limited within the central angle ψ by two arcs of circles, the distance between the latter being 1 cm. Namely

$$\left(\frac{dq}{dt}\right)_\psi = \psi r\,(b^2 - x_1^2 \tau_1) = \psi x_1^2 r\,(\Theta - \tau_1). \tag{21}$$

Let us determine in the same way the amount of heat given away by the atmosphere for the extra-radiation over a similar stripe of the continent.

$$\left(\frac{dq}{dt}\right)_\psi = \psi x_2^2 r \tau_2. \tag{22}$$

According to equations (21) and (22), the branches of the curves of the same name in Fig. 2 have been calculated. They show the changes in the corresponding value with the growth of the distance from the centre of the sea. As it was natural to expect, the curve breaks over the coast line. Under natural conditions the picture must be more smooth: instead of breaking the curve there would cross zero. Such a smooth transition is shown in the corresponding diagram of work (1). Only the crossing of the axis of abscissae by the curve must occur on the coast.

Now let us take a three-dimensional model instead of the two-dimensional one: let us imagine that the heat is transferred by air currents, the velocity of which varies with altitude according to the simple law of sine, as in the cited work (1). The results of the analysis will be easily made more precise by means of the method described in (2). Thus, let the radial component of the wind velocity be expressed by the equation:

$$u = U \sin az; \quad a = \frac{\pi}{D}, \tag{23}$$

where D is the «altitude of friction». In its turn on the ground of the formulae (27) and (73) of the work (1), we can (5) write down

$$U = \frac{\Pi}{2\delta\omega} \cdot \frac{d\tau}{dr}. \tag{24}$$

But in such a case the relations found (23), (24) and (7)—(10) give

$$u_1 = -\frac{\Pi \varkappa_1}{2\delta\omega} N_1 I_1(\varkappa_1 r) \sin az, \tag{25}$$

$$u_2 = -\frac{\Pi \varkappa_2}{2\delta\omega} N_2 K_1(\varkappa_2 r) \sin az. \tag{26}$$

The vertical components of the wind will be determined from the condition of continuity

$$\frac{1}{r}\frac{d(ru)}{dr} + \frac{dw}{dz} = 0.$$

Namely

$$w_1 = \frac{\Pi \varkappa_1^2 D}{2\pi\delta\omega} N_1 I_0(\varkappa_1 r)[1 - \cos az]. \tag{27}$$

$$w_2 = -\frac{\Pi \varkappa_2^2 D}{2\pi\delta\omega} N_2 K_0(\varkappa_2 r)[1 - \cos az]. \tag{28}$$

The curves U and W representing the changes of the radial and vertical components with the growth of distance from the centre of the sea are drawn in Fig. 2.

The value U has the same meaning as in (24). The value W is a function standing as a factor before the square bracket in (27) and (28). Of course under natural conditions the curve U is rather smoothed near the peak, and the curve W crosses zero without a break. The scales of the ordinates of both curves in the draught are different.

On the ground of formulae (25)—(28), it is easy to find the equations of the projections of the stream lines on the vertical radial plane. For the branches lying over the sea or the continent the equations will be:

$$z_1 = \frac{D}{\pi} \cdot \arccos\left[1 - \frac{C_1}{rI_1(\varkappa_1 r)}\right], \tag{29}$$

$$z_2 = \frac{D}{\pi} \cdot \arccos\left[1 - \frac{C_2}{rK_1(\varkappa_2 r)}\right]. \tag{30}$$

The projections of the current lines drawn respectively to equations (28), (30) are represented in Fig. 3. It is remarkable that the picture obtained is intermediate between the two variations of stream lines found in work [1]. It is nearer the variant which was taken as the most satisfactory in the work cited. This once more proves the validity of both our methods when investigating the monsoon field. The breaks between the branches of the curves seen in Fig. 3 are smoothed under natural conditions owing to the above-mentioned reasons.

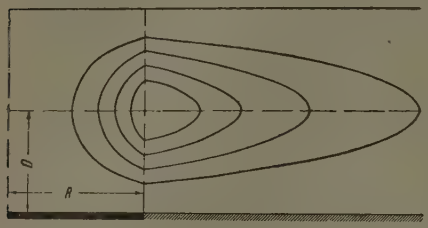

Fig. 3.

At last we must note that along with unlimited growth of the sea radius the velocity of the monsoon wind on the shore tends to a definite limit. Conformably to the coast line the limit value U_∞ will have the expression

$$U_\infty = \frac{\pi\Theta}{2\delta\omega}\left(\frac{\varkappa_1\varkappa_2}{\varkappa_1+\varkappa_2}\right). \tag{31}$$

In its turn by means of this maximum value it is easy to express the value U_R corresponding to certain dimensions of the sea (or island)

$$\frac{U_R}{U_\infty} = \frac{1+\mu}{\dfrac{K_0(\varkappa_2 R)}{K_1(\varkappa_2 R)} + \mu\dfrac{I_0(\varkappa_1 R)}{I_1(\varkappa_1 R)}} \tag{32}$$

The law of the increase of $\frac{U_R}{U_\infty}$ with the increase of the dimension of the sea or island is represented in Fig. 1. The curve *5* corresponds to the first object, the *6* — to the second.

It is obvious that with unlimited growth of $\varkappa_1 R$, the ordinates of both curves tend to 1, which is quite natural, taking into account both physical considerations and the symmetry of formula (31) with respect to $\varkappa_1$ and $\varkappa_2$. It should be noticed that the calculation of the numerical value of U_∞ by means of formula (31) gives the order of magnitude of about 18—20 meters per second.

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GEOPHYSICS

A MORE PRECISE DETERMINATION OF HUMIDITY FROM THE RECORDS OF AEROLOGICAL SOUNDS

By W. W. SHOULEJKIN, Corresponding Member of the Academy

Up to the present time the recording of humidity at various altitudes above the sea is made by means of hygrographs with hairs. Until a new and more perfect apparatus is invented, it is necessary to eliminate in some way a quite substantial error common to all generally used hygrographs. It is known that this error consists in the following: a somewhat precise and reliable record of humidity would be possible under the condition of an extremely low ascension of the aerological sound; with the existing velocities of ascension the apparatus crosses the strata with varying humidity too rapidly and therefore its hair has no time to assume a stabile state and practically we do not known what it records.

Under rapidly varying humidity and alternating wetting and drying of the hair the records undergo the extremely harmful influence not only of the specific sorption-inertia of the hair, but also a pronounced hysteresis (though the latter leads to lesser errors): the velocity of the variations of the hair length under wetting and drying are not equal.

In the present article we shall try to suggest a method by means of which the records of hair hygrographs may be deciphered, allowance being made for the unstationary regime of the apparatus operation.

Let us assume that a hair hygrograph has been kept at a certain original humidity of the air for a long time, after which it is transferred into the air of different humidity. Then we can write down that the original state of the hair will differ from the final S by a certain value Δ which can have the sign—or+.

We have sufficient grounds to suppose that the transition from the original state of the hair to the final one will proceed according to the simple law

$$A = S + \Delta e^{-kt}. \qquad (1)$$

At each given moment of time t the hygrograph will record its reading A corresponding to the transitional regime. These values A are written on all the tapes of the hygrographs in all the ascensions at all times.

Proceeding however from the equation (1) which can be verified experimentally and has been verified by V. G. Dybchenko, it is easy to connect the imaginary value A with the value S corresponding to the true humidity in the stratum crossed.

Indeed, the variable term of the right part (1) can be easily expressed by means of the derivative in time $\frac{dA}{dt}$

$$\Delta e^{-kt} = \frac{1}{k} \cdot \frac{dA}{dt} . \tag{2}$$

But in such a case obviously

$$S = A + \frac{1}{k} \cdot \frac{dA}{dt} . \tag{3}$$

Thus, the true humidity of air corresponding to S is determined according to the fictitious «humidity» which has been recorded by the hygrographs and the velocity of the variation of this fictitious «humidity» in time.

It is quite necessary to note that the value of the parameter k making part of the formula (3) must depend upon: a) the temperature of the air, b) the direction of the process, i. e. on whether the hygrograph passes from the more humid stratum into a less humide one or the opposite. None of these circumstances can bring any difficulty into the treating of the records: it will be necessary only to work out corresponding tables or to construct very simple devices with netted transparents.

There are reasons to suppose that the parameter k depends also on the sought for value S itself, though this dependence is probably very slightly expressed. However, it is easy to prove that even in the case if this dependence will have to be considered, under the existing not too strict rates of accuracy in the measurements of humidity, the correlation (3) will allow to determine the real humidity corresponding to value S by means of the fictitious value A taken from the tape of the recorder.

Indeed let us suppose that the dependence of k on S is expressed by some complex function. Let us suppose that in experimental determination it was impossible to express it analytically, and we could only draw a corresponding graph. The most convenient form for this graph will be the following: the values $\frac{1}{k}$ are put along the axis of abscissae and S along the axis of ordinates (curve *1*).

But on the same diagram we may represent the curve *2* plotted according to equation (3). Then the ordinate of the crossing of both curves will immediately represent the sought for value S.

Of course, in practice it will be possible to avoid the plotting of similar curves for each case: the construction of a simple device (with a netted transparent and a rotating ruler), which will allow to determine S automatically by imposition on the hygrograph tapes, suggests itself.

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CHEMICAL PHYSICS

ON THE TRANSITION OF THE COMBUSTION OF EXPLOSIVES INTO DETONATION

By J. B. CHARITON and B. M. STEPANOV

(Communicated by N. N. Semenov, Member of the Academy, 10. III. 1939)

Cases are known when the combustion of an explosive developed into a sudden explosion, accompanied by a violent mechanical action, the nature of which suggests that a detonation process must have taken place.

The literature on the subject contains conclusions of various authors on the conditions necessary for the transition from combustion to detonation; however, we did not come across any experimental paper relating to the phenomenon itself. The present research was undertaken with the aim of obtaining some quantitative data concerning the transition from combustion to detonation.

Highly sensitive explosive substances were first investigated, such as methylnitrate, nitroglycol, nitroglycerin, explosive mixtures of methylnitrate and ether, methyl alcohol or nitrobenzene, as well as explosive gelatins consisting of a methylnitrate base with varying contents of pyroxylin. The explosive substance was poured into a metallic container 7 mm in diameter placed in a thick-walled bomb (28 cm^3). The ignition of the explosive was effected with the aid of some gun-cotton which in its turn was ignited by means of an electrically heated wire. Detonation, or a regime akin to detonation, manifested itself by the destruction of the container holding the explosive. In the case of combustion the container remained entirely undamaged; as soon as detonation set in, the bottom of the container was knocked out and other mechanical damage occurred.

In the course of our experiments we found that, up to densities of 0.009 g per cm^3, methylnitrate, when ignited in a closed bomb, undergoes combustion without any mechanical action. At densities above this figure, the combustion of the methylnitrate always develops into detonation, the destructive effect being the more considerable, the greater the density. Experiments with nitroglycol and nitroglycerin showed the combustion to proceed, while the density is below 0.015 g per cm^3, developing into detonation at higher densities*. However, the results for nitroglycol and nitroglycerin cannot be regarded as conclusive; both substances are not easily inflammable, and some explosive gelatin had to be introduced to ignite them. It might there-

* It should be noted that in some cases detonation was obtained even at densities of 0.01 g nitroglycol per cm^3, the mechanical action being, however, somewhat less pronounced than with the same amount of methylnitrate.

fore be that the small amount of explosive substance usually dealt with in our experiments became heated, while the combustion of the explosive gelatin was going on, so that the combustion of the substance under investigation proceeded at a higher temperature, which facilitated the transition from combustion to detonation (1).

Mixtures of methylnitrate and nitroglycerin behave very much like methylnitrate or nitroglycerin alone. At densities of 0.0072 g per cm^3 a mixture of 90% methylnitrate and 10% nitroglycerin undergoes combustion, while at greater densities the combustion invariably develops into detonation. The fact that in the case of the above mixture combustion passes to detonation at a somewhat lower density than in that of methylnitrate alone, might be possibly explained by the positive oxygen balance for nitroglycerin compared with the negative balance for methylnitrate, so that the excess of oxygen remaining after the combustion of the nitroglycerin will be consumed in the combustion of the methylnitrate, thus leading to an increase of the combustion temperature of the mixture.

For a mixture consisting of 50% methylnitrate and 50% nitroglycerin, combustion passes to detonation at a density of 0.009 g per cm^3 and over, just as in the case of methylnitrate alone.

Upon addition of inert substances to the mixture, the transition to detonation proceeds less readily. In the table below are summarized the results of a few experiments; under the heading Δ_c (critical density) is given the minimum density, at which the combustion of the corresponding explosive mixture begins developing into detonation.

Composition of the mixture		Δ_c	Composition of the mixture		Δ_c	Composition of the mixture		Δ_c
Methylnitrate, %	Ether, %		Methylnitrate, %	Methylalcohol, %		Methylnitrate, %	Nitrobenzene, %	
95	5	0.009	—	—	—	—	—	—
90	10	0.0143	90	10	0.0107	—	—	—
80	20	0.0375	80	20	0.0357	80	20	0.0143

It is easily seen that the addition of an inert substance hinders the transition from combustion to detonation. Given the same percentage of the admixture, in the case of methylnitrate the transition from combustion to detonation proceeds the less readily, the greater the vapour pressure of the inert admixture. The above rule (if a rule can be established on the basis of not more than three different admixtures) confirms the view expressed by Belajev(2), according to which, other conditions being the same, combustion develops into detonation the more readily, the less volatile the given explosive.

Experiments conducted with explosive gelatins (methylnitrate plus pyroxylin) yielded the following results: the minimum density, at which the combustion of a mixture of 99.5% methylnitrate and 0.5% pyroxylin passes to detonation, was found equal to 0.009 g per cm^3, just as in the case of methylnitrate alone. An increase of the pyroxylin content to 1% markedly affected the minimum density, at which the combustion developed into detonation, which in this case increased to $\Delta_c=0.1$ g per cm^3, while a 20% content

of an inert liquid was found to raise the minimum density required for detonation only to 0.04 g per cm^3. It is quite obvious that the physical state of the admixture plays an important part in the process. Indeed, while the explosive gelatin prepared by adding 1% of pyroxylin to the methylnitrate presents a jelly-like mass, any substance containing inert admixtures is a liquid of very low viscosity, a mixture of methylnitrate and 0.5% pyroxylin being a viscous liquid. The influence of the physical state on the conditions for the transition from combustion to detonation is probably closely connected with the well-known fact that the sensitivity of an explosive to the initiating impulse depends on its physical state.

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CHEMICAL PHYSICS

ON THE TRANSMISSION OF DETONATION FROM GASES TO EXPLOSIVES

By J. B. CHARITON and E. V. RDULTOVSKAJA

(Communicated by N. N. Semenov, Member of the Academy, 10. III. 1939)

When the detonation is transmitted from one explosive to another, in addition to the effect of the detonation wave proper, the high-speed particles ejected by the detonating substance may also play an important part. The quantitative investigation of the conditions causing detonation of explosives is therefore best carried out with an explosive gas as the source of the detonation wave. Explosive gases present the additional advantage that, by varying the pressure, the amplitude of the detonation wave can also be varied to a very wide extent. In the case of solid or liquid explosives, the amplitude of the detonation wave can be varied only within very narrow limits.

In the present paper a few data are given on the transmission of the detonation started in a stoichiometric hydrogen-oxygen mixture to explosives.

The experiments were conducted in the following way. A 100 cm long steel cylinder having an inner diameter of 23 mm was filled with a mixture of $2H_2+O_2$ from an electrolyzer, to which it was connected by means of a valve and a copper tube. The pressure of the mixture was proportional to the amount of electricity passed through the electrolyzer and was read on a manometer, which was also connected to the cylinder by means of a valve. A cup containing the explosive under investigation was placed in the lower part of the cylinder. The upper part was provided with a special sparking device for the ignition of the mixture. Some preliminary experiments showed that the detonation in the upper part of the cylinder started after the flame had travelled some 40 cm.

Prior to filling the cylinder and the electrolyzer with the mixture to be investigated, the air was removed by means of an oil pump.

In the first place the transmission of the detonation to nitroglycerin and methylnitrate was studied.

So long as the initial pressure of the mixture $2H_2+O_2$ did not exceed 6 atm., the nitroglycerin in the cup remained unchanged. At pressures higher than this, no nitroglycerin remained in the cup.

In order to verify whether the disappearance of the nitroglycerin should not be ascribed to evaporation, it was substituted by water. After the detonation, the water was still found in the cup. Since the vapour pressure of nitroglycerin is less than that of water, no evaporation could have taken place in the first case, and we are led to assume a combustion process. On a further

increase of the pressure up to 27 atm., no other phenomena could be observed.

In the case of methylnitrate, combustion was observed for pressures up to 10 atm. At 14 atm., however, detonation of the mixture caused detonation of the methylnitrate.

It will be of interest to compare the above results with those reported by Chariton and Stepanov in the foregoing paper on the transition from combustion to detonation. The combustion of methylnitrate was found then to change to detonation at densities about 0.01 g per cm^3, i. e. at pressures of about 100 atm. Now, in the case of a mixture of $2H_2+O_2$, after its explosion the pressure attained some 300 atm.

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CHEMISTRY

COMPLEX COMPOUNDS OF PLATINUM AND BUTADIENE (DIVINYL)

By ANNA GELMAN

(Communicated by N. S. Kurnakov, Member of the Academy, 28. III. 1939)

A study of complex compounds of platinum and ethylene and their homologues (1) showed that these hydrocarbons occupy only one co-ordination place in full accord with Chugayev's cycle rule (2).

The presence of two double bonds in butadiene made us assume that it could occupy two co-ordination places, forming a stable five-member cycle. This provokes interest in the respect that up till now we have not been successful in establishing whether a central atom can co-ordinate two bonds at all, for all our attempts to produce compounds of the type $Pt(CnH_{2n})_2Cl_2$ were a failure. Besides that it was of interest to establish: 1) whether both double bonds of butadiene are always used up in the formation of complexes; 2) the trans-influence of these bonds; 3) a space isomery attainable in the butadiene complexes.

When an acidified aqueous chloroplatinite solution was treated with butadiene, the solution turned yellow after two days. After being separated from a scant brown precipitate, the transparent bright yellow filtrate was divided into two parts. One part was slowly evaporated in air, as a result of which splendid orange-yellow needles precipitated. The crystals were filtered through a Büchner funnel, washed with alcohol and benzene and dried over calcium chloride.

Reaction went on according to the following equation:

$$2\,(NH_4)_2\,PtCl_4 + CH_2 = CH - CH = CH_2 \rightarrow \begin{matrix} NH_4 \\ NH_4 \end{matrix}\left[\begin{matrix} Cl \\ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} C_4H_6 \\ Cl\ \ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} Cl \\ Cl \end{matrix}\right] + 2NH_4Cl. \qquad (I)$$

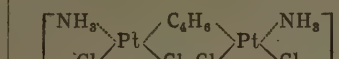
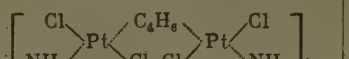
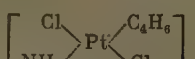
Substance I reacts with the salt of Reise's first base according to the equation:

$$\begin{matrix} NH_4 \\ NH_4 \end{matrix}\left[\begin{matrix} Cl \\ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} C_4H_6 \\ Cl\ \ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} Cl \\ Cl \end{matrix}\right] + (NH_3)_4\,PtCl_2 \rightarrow$$

$$\rightarrow [(NH_3)_4\,Pt]\left[\begin{matrix} Cl \\ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} C_4H_6 \\ Cl\ \ Cl \end{matrix}\!\!>\!Pt<\!\!\begin{matrix} Cl \\ Cl \end{matrix}\right] + 2NH_4Cl. \qquad (II)$$

Ammonia was added drop by drop to the second part of the solution which was stirred constantly with a glass stirring rod. A yellow-coloured precipitate appeared immediately, but disappeared when stirred. After neutralizing a fine crystalline ochre-yellow precipitate appeared.

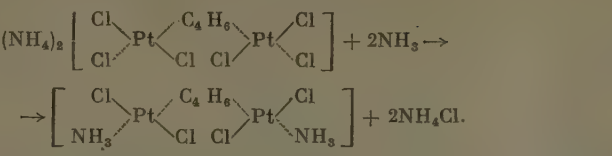
Table 1

Formula of compound	 IV	 III	 VIII
Colour	Pale yellow	Ochre-yellow	Dirty yellow
Optic * constants	Under the microscope the substance is finely crystalline. Is not subject to optic determination	Under the microscope the substance is isotropic, fine-grained $N = 1.77—1.78$	Not determined
Electroconductivity at 25°C **	$\rho = 10\,000$ $\mu = 279.02$ 24h.	Dissolves poorly, decomposing. We were unable to determine μ	Not determined
Reaction with $AgNO_3$ in the cold	AgCl precipitates immediately	No change observed for 10 min.	Slight opalescence appears after about 10 min.
Reaction with HCl	Insoluble	Dissolves forming $(NH_4)_2(PtCl_3)_2C_4H_6$	Dissolves
Reaction with thiourea	Dissolves forming $Pt4ThCl_2$	Ditto	Ditto
Reaction in hot water	Slowly decomposes liberating platinum	Decomposes	Decomposes
Reaction with NaOH	Decomposes	Ditto	Ditto
Result of heating in a capillary tube in air	Decomposes without melting at 202—204°C	Decomposes without melting at 185—188°C	Melts and decomposes at 172—174°C

* For determination of electroconductivity see M. M. Yakshin's paper and optic constants—M. N. Lyashenko.

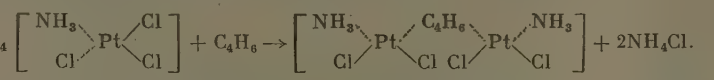
** The determination was made from a test of a solution containing some remains of the precipitate, since the substance is very slightly soluble.

The equation of the reaction is as follows:



(III)

When an acidified solution of Kossa's ammonium salt was treated with divinyl, a pale yellow crystalline precipitate appeared after 24 hours. The reaction expected was:

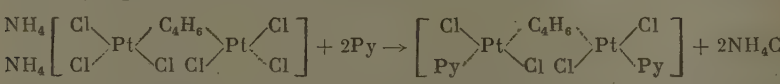


(IV)

An analysis fully confirmed our assumptions.

We give III a trans and IV a cis-configuration according to their analogy with the ethylene compounds, since III was produced from a salt of the Zeiss type, and IV from Kossa's salt (3). This follows from an examination of the properties of these compounds (Table 1).

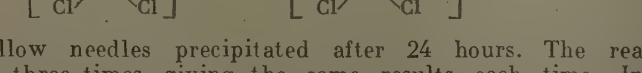
Butadiene-pyridine platinum chloride was produced according to the following equation:



(V)

This orange-yellow, finely crystalline substance is entirely similar in properties to III (Table 2).

When an acidified solution of Kossa's pyridine salt was treated with butadiene the reaction went on according to the equation:



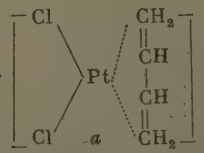
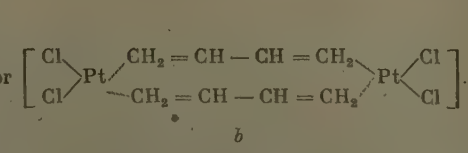
(VI)

the pale yellow needles precipitated after 24 hours. The reaction was repeated three times, giving the same results each time. In this case butadiene combines by means of only one double bond.

We were unsuccessful in producing the cis-isomer for V, but instead we were successful in obtaining the trans-isomer for compound VI under the following conditions.

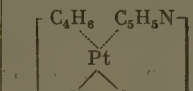
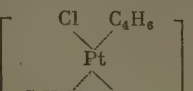
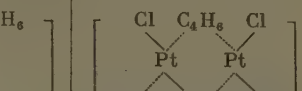
In the beginning of this paper we have already mentioned that when chloroplatinite was treated with butadiene a small brown-coloured precipitate appeared. In neutral solution the same reaction resulted in the precipitation of a considerably more abundant greenish-brown precipitate.

What compound could have formed under these conditions? Two variants are possible:

either  *a* or  *b*.

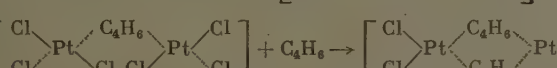
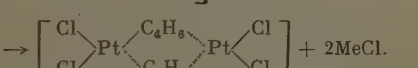
I am, however, inclined to accept formula *b* for this compound. Its formation seems to be the second phase of the reaction between

Table 2

Formula of compound	 VI	 VII	 V
Colour	Straw-yellow	Yellow	Orange-yellow
Electroconductivity at 25°C	10000 μ 98.7; 116.9; 135.0 t_{min} = 30; 40; 50	Could not be determined as a result of very poor solubility μ	Not determined
Reaction with $AgNO_3$ in the cold	Opalescence appeared immediately and AgCl precipitates	Weak opalescence, no precipitate	Weak opalescence
Reaction with HCl in the cold	Does not dissolve	Dissolves forming $PyH(PtC_4H_6Cl_3)$	Dissolves forming $(PyH)_2(PtCl_3)_2C_4H_6$
Reaction with thiourea	Dissolves in the cold, forming $Pt4ThCl_2$	Ditto	Ditto
Reaction in hot water	Decomposes slowly	Decomposes	Decomposes
Result of heating in a capillary tube in air	Changes colour at 130°, melts and decomposes at 155—158°	Melts at 125—130°, decomposes at 130—135°	Melts at 138—140°, decomposes at about 145°

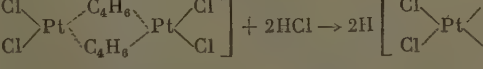
butadiene and chloroplatinite:

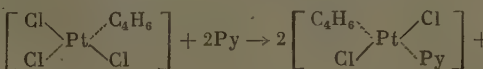
1. $2Me_2PtCl_4 + C_4H_6 \rightarrow Me_2$ $+ 2MeCl.$

2. Me_2  $+ C_4H_6 \rightarrow$  $+ 2MeCl.$

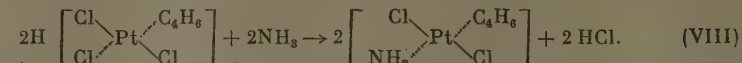
The greenish-brown substance dissolves in dilute hydrochloric acid when heated. When this is done, there is a strong smell of butadiene and a brownish precipitate appears.

After filtering off the pricipitate we got a transparent orange-yellow solution. After the solution had been cooled, pyridine precipitated a yellow fine crystalline substance. It dissolved easily in hydrochloric acid and was reprecipitated with pyridine. The equations of these reactions may be represented in the following form:

1.  $+ 2HCl \rightarrow 2H$ (VII)

2. $2H$  $+ 2Py \rightarrow 2$ $+ 2HCl.$

The reaction with ammonia goes on just as smoothly:

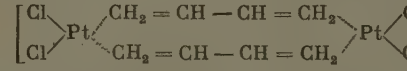


(VIII)

Passing over to a discussion of the experimental material, it must be mentioned first of all that both double bonds of the butadiene molecule act separately in co-ordinational addition.

Butadiene forms a kind of bridge between the two c. a., giving compounds I, II, III, IV, V. Each of the butadiene double bonds shows a high trans-influence, just as has been observed in ethylene and its homologues.

Butadiene apparently does not give a five-member cycle with c. a. Most probably a cycle of the following type is formed.



It is easily broken up with hydrochloric acid, forming $H[PtC_4H_6Cl_3]$.

The fact that butadiene does not close a cycle with one c. a. can be explained through steric obstacles — the absence of free rotation in the butadiene molecule as a result of the proximity of the two double bonds. On the other hand, it is quite possible that the c. a. co-ordinates stably only with one double bond. In this sense it would be interesting to synthesize complex compounds with diallyl, in which the double bonds are apart.

The behaviour of butadiene compounds is exactly the same as that of the corresponding ethylene complex platinum compounds.

Conclusions

1. Nine butadiene complex platinum compounds have been synthesized.

2. It has been established that either one or both double bonds of the butadiene molecule may take part in co-ordinational addition.

3. It has been established that each double butadiene bond has a high trans-activity within the interior sphere of the complex.

4. Space isomeres of platinum butadieneamindichlorides have been produced by installation in turn of neutral molecules.

The study is in progress.

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Academy of Sciences of the USSR.

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31. III. 1939.

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CHEMISTRY

OXIDATION OF CuS AND TRANSFERENCE OF Cu INTO AQUEOUS SOLUTION AT AIR PRESSURE

By O. E. ZVIAGINCEV and V. G. TRONEV

(Communicated by N. S. Kurnakov, Member of the Academy, 29.III.1939)

According to publications, both old experimental, e. g. (1), and new theoretical ones, e. g. (2), the solubility of copper sulphides in water, whether they are natural minerals or artificially precipitated from solutions, fluctuates within the limits of very small values of the order of $1 \cdot 10^{-13}$ to $1 \cdot 10^{-6}$ grammoles per litre at room temperature and atmospheric pressure.

Naturally, as the temperature rises, the solubility of the sulphides in water increases, and Verhoogen's thermodynamical calculations(2) show that the solubility of the sulphide can be increased many thousand times by raising the temperature to a few hundred degrees.

The results of Cohen's work (4) show that hydraulic pressure has little effect on the solubility of solid substances in liquids, which may be explained by the very small changes brought about by pressure in volume and in the other properties of solid and liquid substances. In the case when there is in the system a gas capable of reacting with the components of the system, the problem of the effect of pressure on the solubility of solid substances and liquids becomes much more complicated and has almost not been studied at all. The effect of pressure on the solubility of a solid substance, copper sulphide for instance, in water in the presence of oxygen, which is capable of oxidizing the sulphide to the sulphate, is determined virtually by the effect of pressure in the gaseous phase on the velocity of the chemical process. There are almost no data published on this kind of systems, since the data on the influence of hydraulic pressure on the reaction velocity in condensed systems, for instance, ether saponification and other such systems, when there is no gaseous phase (these data are in themselves contradictory), cannot give any indications as to the behaviour of sulphides in water under air pressure.

However, in analogy to the results arrived at in the previous works of Tronev and Bondin (5) on the effect of air pressure on the solution of noble metals—platinum in acids and gold in cyanides, it may be assumed that air pressure favourably affects the solution of sulphides in water and other solvents in connexion with the possibility of the sulphides being oxidized to soluble sulphates, especially if the temperature is raised at the same time above 100°.

In this report we give preliminary data on the effect of high air pressure (up to 100 atm) and temperature (up to 150°) on oxidation and solution in water.

1. Effect of Partial Pressure of Oxygen on the Solution of Cu from CuS in Water. For the experiments we took a weighed portion of copper sulphide, precipitated from a $CuSO_4$ solution with hydrogen sulphide and dried at 100° to constant weight.

While recurring to the electroconductivity method in determining the solubility in distilled water of our CuS sample under usual conditions of oxidation, temperature and air pressure, we obtained data which agree satisfactorily enough with the aqueous solubility values obtained by Weigel[1], but conflict with those obtained by Verhoogen[2].

According to Weigel for 24 hours $3.51 \cdot 10^{-6}$ g moles per l; according to Verhoogen (calc. at 25°) $2.4 \cdot 10^{-13}$ g moles per l; our sample $5 \cdot 10^{-6}$g moles per l.

A weighed portion of the initial material was put into a glass beaker with 100 ml distilled water. The beaker was put into the usual high pressure autoclave with a glass stirring rod, whose cap covered the opening of the beaker with the weighed portion non-hermetically in order to keep out foreign admixtures (oil from the stuffing-box, corrosion products from the material of the autoclave etc.). Then the autoclave was closed, placed in an oil thermostat furnished with transmission and a thermoregulator, was filled with air or a nitrogen-oxygen mixture to the required pressure and was kept thus for a definite length of time, the temperature and pressure conditions and stirring velocity being kept accurately constant. After the experiment the solid and liquid phases were analyzed for copper and sulphur content in the usual manner.

The results of the experiments for various partial pressures of oxygen in the gaseous phase are given in Table 1. This table shows that as the partial pressure of oxygen rises, the amount of CuS dissolved in water at 100° quickly increases. It was found that the solution contained copper sulphate and the precipitate was a mixture of the CuS which had not taken part in the reaction and the hydrolysis products of $CuSO_4$ in the form of a green-coloured precipitate.

Table 1

Constant experiment conditions: weighed portion of CuS—1 g; 100 ml distilled water; duration of experiments—2 hours; temperature—100°; total pressure in the gaseous phase without heating—100 atm.

Partial pressure of oxygen in the system in atm.	Quantity of copper in g dissolved in 100 ml H_2O in the form of $CuSO_4$	Per cent Cu dissolved	Notes
0.3	0.04	6.0	Commercial nitrogen with an admixture of C_2 was used
16	0.30	45.4	Artificial mixture
17	0.31	47.0	Ditto
20	0.33	50.0	Air
46	0.58	87.9	Mixture of nitrogen and O_2
46	0.58	87.9	Ditto. Experiment lasted 6 hrs instead of 2 hrs.

The last, sixth experiment which was conducted a longer time (6 hours instead of 2 hours, as in the previous experiments) shows that both the solution velocity and the quantity of Cu which passes into aqueous solution reach their limit in 2 hours. Evidently, as the main CuS mass becomes oxidized to $CuSO_4$, the latter hydrolizes forming basic copper sulphate which is insoluble in water as a result of which we get an equilibrium of the following type:

$$3CuSO_4 + 4H_2O \rightleftarrows CuSO_4 2Cu(OH)_2 + 2H_2SO_4.$$

As the concentration of $CuSO_4$ increases with the oxidation of CuS, the equilibrium shifts towards the right, where crystals of the basic salt are precipitated. We were unsuccessful in shifting this equilibrium considerably towards the solution of CuS by increasing the time (up to 24 hours), since the last CuS particles become covered with hydrolysis products and pass into $CuSO_4$ very slowly.

An analysis of the green precipitate thus produced through the oxidation of CuS, and also in a separate experiment through the hydrolysis of a saturated $CuSO_4$ solution at 100°, gave the following results:

I. The basic salt produced through the oxidation of CuS contains Cu—53.9%; SO_4—27.0%.

II. The basic salt produced by hydrolizing $CuSO_4$: Cu—52.2%; SO_4—29.4%.

According to published data the green precipitate of basic copper sulphate has the following composition. From theory: Cu—53.7%; SO_4—27.07%; from Schenstone's data([6]) (produced by hydrolizing $CuSO_4$): Cu—54.90%, 54.08%; SO_4—26.78%, 27.18%.

Analogous results for the composition of basic copper sulphate were obtained by Binder ([7]) and Genesley([8]) who produced the crystals under other conditions.

Thus, along with the explanation of the problem of the effect of pressure, which greatly increases the solution of CuS in water, the mechanism of the processes which take place under these conditions also becomes clear, viz.:

$$CuS + 2O_2 = CuSO_4. \quad (I)$$

$$3CuSO_4 + 4H_2O \rightleftarrows CuSO_4 2Cu(OH)_2 + 2H_2SO_4. \quad (II)$$

II. Influence of Temperature on the Solution of Cu from CuS in Distilled Water under Air Pressure. The next series of experiments with varying temperatures and other conditions constant was performed in order to confirm the mechanism of CuS oxidation proposed and to establish the relation between the velocity with which Cu passes into aqueous solution and the collateral hydrolysis process which is evidently connected with the equilibrium mentioned above (II). The results of the experiments are given in Table 2.

The experiment results in Table 2 show that first of all the CuS oxidation velocity and the passing of Cu into solution increases as the temperature is raised until an optimum is reached, above which solution decreases more or less pronouncedly depending on the initial weighed portion of CuS, which may be explained in terms of the acceleration and strengthening of hydrolysis of the sulphate product. The formation of considerable quantities of free sulphuric acid in the solution during the experiments of dissolving CuS at temperatures 150, 200 and 250° shows that there is an equilibrous state at each temperature between the oxidation products of CuS—$CuSO_4$ and the products of the hydrolytic decomposition of the latter—the basic salt and sulphuric acid. We shall return to the problem of these equilibrious states and their study by measuring their electroconductivity at high pressures and temperatures in one of our future papers.

Table 2

Experiment conditions: air pressure 100 atm. (without heating); duration 2 hours; volume of solvent 100 ml; stirring was applied

Weighed portion of CuS, g	Temperature	Dissolved in 100 ml			Notes
		Cu, g	SO_4, g	Cu, %	
20	100	0.66	none	10.0	Considerable embedment of unreacted CuS in the basic salt mass
10	150	1.22	1.5	15.4	
10	200	1.23	2.99	18.6	
10	250	0.17	1.51	2.5	
1	100	0.33	—	50 0	
1	150	0.56	—	84.8	
1	200	0.61	—	92.4	
1	250	0.08	—	12.0	

Conclusions

1. Copper sulphide can be dissolved in distilled water in a few hours under air pressure at temperatures from 100 to 200°. Copper sulphates are formed.

2. The velocity with which Cu passes into aqueous solution is proportional to partial pressure of oxygen in the gaseous phase.

3. As a result of hydrolysis of the copper sulphate which forms from the oxidation of CuS, basic copper sulphate $CuSO_4\,2\,Cu\,(OH)_2$ remains undissolved in water. Its quantity depends upon the temperature.

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CHEMISTRY

OXIDATION OF ZnS AND TRANSFERENCE OF Zn INTO AQUEOUS OR ALKALI SOLUTION AT AIR PRESSURE

By V. G. TRONEV and S. M. BONDIN

(Communicated by N. S. Kurnakov, Member of the Academy, 29.III.1939)

For reasons set forth in the first communication by Zviagincev and Tronev(1) on the problem of transference of copper sulphide at high pressures into solution in water, it was of some interest to study, likewise, the influence of the pressure of air on the velocity and the mechanism of dissolving of zinc sulphide in water or certain other solvents.

There are no experimental data to be found in the literature on the dissolving of ZnS in water, diluted acids and alkalies at the temperatures above 100° and, consequently, at high pressures. The solubility of ZnS in water is quite insignificant under ordinary conditions; according to Weigel(2), it amounts to $71.6 \cdot 10^{-6}$ grammoles per liter for the salt precipitated from solutions and to $6.65 \cdot 10^{-6}$ grammoles per liter for the natural mineral sphalerite. In 1938 Verhoogen(3) made thermodynamic calculations of the dependence of the solubility of various sulphides and, in particular, of ZnS on the temperature in the range from 25 to 400° and, likewise, on the concentration of hydrogen and hydroxyl ions in the solvent.

Artificial zinc sulphide, prepared by the method of Vanino by precipitating it with ammonium sulphide from a solution of zinc acetate and dried at 100°, served as an initial material.

The experiments were carried out in a common autoclave of 1 l capacity with a stirrer. In case of non-alkaline solutions a glass beaker and a glass screw-formed stirrer were placed in the autoclave for the purpose, whereas the experiments with alkaline solution were carried out in the steel beaker of the autoclave itself. The autoclave was placed, as usual, in a thermostat provided with a thermoregulator, transmission and other adjustments and kept for a certain period at an adequate pressure of nitrogen or air. Samples to be analyzed for the content of zinc and sulphur in the solution and the solid phase were drawn from the autoclave after it had been cooled or by means of certain adjustment taken in given intervals of time, while the autoclave was being heated. Zn was determined as pyrophosphate or by electrolysis after the method of Amberg(4) or Spitzer(5). The sulphur was determined in the form of $BaSO_4$ from hydrochloric solution.

First experiments which were of preliminary nature had for object to ascertain the rôle of the solvent for ZnS in the range of temperatures from 100 to 200° at air pressure (Table 1).

Table 1

Influence of the Solvent on the Rate of Oxidation and Transference of ZnS into Solution at Temperatures from 100 to 200° at Air Pressure (Experimental conditions: the weight of the sample was 1.5—2 g; the volume of the solvent 100 ml; the experiment lasted 2 hours; the agitation was carried out with the rate of 200 revs. per hour)

Solvent	Pressure of air, atm.	Temperature	Weight of sample, g	Found ZnS in g per 100 ml of solution
H_2O	1	100°	1.8723	0.0004
H_2O	85	100	1.4865	0.0016
H_2O	80	200	1.6732	0.0048
0.1 *N* sol. H_2SO_4 . . .	85	100	1.8131	0.0104
0.1 *N* sol. H_2SO_4 . . .	1	100	1.5111	0.0069
4 *N* sol. NaOH	80	150	0.6891	0.3976

The following conclusions can be drawn from the data given in Table 1.

1. The high pressure of air (up to 85 atm.) compared with atmospheric pressure increases, other conditions being equal, the transference of Zn into aqueous solution by four times.

2. The increase of temperature up to 100° at air pressure of 85 atm. increases considerably the transference of Zn into the solution, as compared with the solubility value at 25°, approaching the data calculated by Verhoogen (from the values of the order of $1 \cdot 10^{-6}$ up to $1 \cdot 10^{-4}$ grammol per liter).

3. The further increase of the temperature from 100 to 200° increases the transference of Zn into solution by three times.

4. As it was expected, the transference of Zn into solution in a strongly diluted acid was not increased considerably by the use of pressures up to 85 atm. at 100° (from 0.0069 g of Zn per 100 ml of water at atmospheric pressure to 0.0104 g at air pressure of 85 atm.).

5. The dissolving of Zn in an alkaline solution was considerably increased at air pressure of 80 atm. and at 150° C.

The next problem in which we were interested, was that of the influence of the pressure of air on the transference of ZnS into solution in a solvent most suitable for swift dissolving (within two hours), namely in an aqueous solution of sodium hydroxide. The results of the experiments at various pressures up to 125 atm. are given in Table 2.

Table 2

Experimental conditions: 100 ml 4 *N* solution of NaOH; the temperature—150°; the duration of the experiments—2 hours with stirring

Sample of ZnS, g	Air pressure, atm.	ZnS transferred into solution, g
1.5220	1	0.2820
2.0601	25	0.4538
1.9231	50	0.7321
1.0403	75	0.9684
1.9278	100	1.2746
1.9829	125	1.2789

From the data given in Table 2 it may be seen that with a 4 *N* solution of NaOH the transference of Zn into the solution increases with the increase of pressure up to 75 or 100 atm.; with the further increase in pressure, the influence of the latter decreases, this phenomenon may be explained on the basis of similar facts observed previously in the authors' earlier studies[6] on the transference into solution at pressure, i. e. by assuming that in an alkaline solution at the pressures from 75 to 100 atm. there sets up a concentration of dissolved oxygen sufficient to oxidize ZnS.

Judging from this nature of the influence of the pressure of air on the transference of ZnS into an alkaline solution, a conjecture might be drawn of the probable mechanism of the process under consideration. This mechanism is connected, apparently, with the primary oxidation of sulphide to sulphite or sulphate with the subsequent formation of sodium zincate according to the following scheme (conjectural):

$$ZnS + 2O_2 = ZnSO_4 \qquad (I)$$

$$ZnSO_4 + 4NaOH = Na_2SO_4 + Na_2ZnO_2 + 2H_2O. \qquad (II)$$

This scheme is corroborated by the presence of the amount of SO_4 in the solution, corresponding with the amount of zinc sulphide transferred into the solution. With a hydrolytic mechanism of dissolving

$$ZnS + 2H_2O \rightleftarrows Zn(OH)_2 + H_2S$$

$$Zn(OH)_2 + 2NaOH \rightleftarrows Na_2ZnO_2 + 2H_2O$$

$$H_2S + 2NaOH = Na_2S + 2H_2O$$

there should have been ion S'' in the solution and this was not detected by the authors. Moreover, the influence of the pressure of air would be difficult to account for, should the hydrolytic scheme be assumed.

Conclusions

1. The insignificant transference of ZnS into solution in water or alkaline solutions under ordinary conditions of temperature and pressure is considerably increased at higher pressures of air. Thus, it is possible to dissolve ZnS completely in a 4 *N* solution of NaOH at a temperature of 150° and air pressure of 100 atm. during a few hours.

2. The process of dissolving of ZnS in alkali is accompanied by its oxidation to $ZnSO_4$ with a subsequent formation of zincate.

Data on the influence of the temperature and the alkali concentration on the solubility of ZnS and other sulphides will be given in the following communications.

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CHEMISTRY

ON MIXED PLATINUM HYDROXYLAMINE TETRAMINES

By V. I. GOREMYKIN and K. A. GLADYSHEVSKAJA

(Communicated by N. S. Kurnakov, Member of the Academy, 29. III. 1939)

In previous papers ([1], [2]) one of us showed how the quantitative ratio of ammonia and pyridine and of hydroxylamine and pyridine in the complex cation affects the properties of the complex compound. In this work we attempted to synthesize compounds, which simultaneously contained three different amines: hydroxylamine, ammonia and pyridine, which differ in composition and structure. The experimental data which we obtained show that it is possible to synthesize tetramines with four different neutral molecules of the $|PtA_1A_2A_3A_4|X_2$ type.

Trans-dihydroxylamine-ammonia-pyridine-platinum chloride $|Pt2HxNH_3Py|Cl_2$. This tetramine is produced by instilling hydroxylamine into the trans non-electrolyte $|PtNH_3PyCl_2| \rightarrow$ which was produced before by Jergensen ([3]), but it was a long time before we were successful in obtaining it pure. Then we made use of an interchange reaction between the chloroplatinite $|Pt2HxNH_3Py|\,|PtCl_4|^* \rightarrow$ and the chloride of Reise's first base, according to the equation:

$$|Pt2HxNH_3Py||PtCl_4| + |Pt4NH_3|Cl_2 \rightarrow |Pt2HxNH_3Py|Cl_2 + |Pt4NH_3||PtCl_4|.$$

The reaction is brought about in the cold in the following manner: the finely ground chloroplatinite is put into a porcelaine evaporating dish, treated with a solution of $|Pt4NH_3|Cl_2$ taken in a quantity 20% less than the theoretical and dissolved in 5 ml water, and is carefully mixed. The reaction goes on quite quickly and is complete in 5—10 min. The precipitate is filtered through a Girsch funnel, and the solution is evaporated in a vacuum exsiccator to 2—3 ml and then is treated with concentrated or better absolute alcohol, which momentaneously precipitates a colourless crystalline substance in the form of thin and sometimes thick needles which dissolve well in water and dilute alcohol. After being dried in the exsiccator over phosphores pentoxide or sulphuric acid, the substance was analyzed. The results of the analysis:

0.0880 g	of substance	gave	0.0400 g Pt
0.0840 »	»	»	0.0384 » »
0.1032 »	»	»	0.0698 g AgCl
0.1086 »	»	»	0.0736 » »

Calculated for $\|Pt2HxNH_3Py\|Cl_2$		Found
% Pt	45.58	45.45; 45.71
% Cl	16.56	16.73; 16.76

* Hx stands for hydroxylamine NH_2OH; Py stands for pyridine C_5H_5N.

The molecular electroconductivity measured at 25°C is in full accord with Werner-Miolati's data for this type of complex.

v	500	1000	2000	4000	8000	16000
μ_v	240	248	254	258	259	264

Trans-chloroplatinite $|Pt2HxNH_3Py|\ |PtCl_4|$. If a solution of the tetramine $|Pt2HxNH_3Py|Cl_2$, acidified with dilute hydrochloric acid, is added to $K_2|PtCl_4|$, an abundant precipitate of a pink crystalline substance in the form of thin needles falls out after 20—30 min. depending upon the concentration of the solution taken. The chloroplatinite can be recrystallized from water by heating gently and shaking often. After being washed a few times with cold water and dilute alcohol and dried at 110° the chloroplatinite gave satisfatory analysis results:

0.1087 g of substance gave 0.0578 g Pt
0.1196 » » » » 0.0672 » »
0.1350 » » » » 0.01106 g AgCl
0.1213 » » » » 0.0986 » »

Calculated for $\|Pt2HxNH_3Py\|\ \|PtCl_4\|$	Found
% Pt 56.24	56.28; 56.19
% Cl 20.43	20.27; 20.11

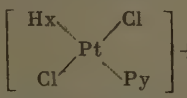
Trans-chloropalladite $|Pt2HxNH_3Py|\ |PdCl_4|$. If the same solution of the tetramine $|Pt2HxNH_3Py|Cl_2$ is treated with a freshly prepared concentrated solution of $K_2|PdCl_4|$, it precipitates a pale lilac-coloured crystalline substance in the form of thin needles which turned out to be non-homogeneous. After this precipitate had been removed by filtering, there appeared a second precipitate reddish-brown in colour and in the form of long tablets.

After being washed with concentrated alcohol and dried in the exsiccator over sulphuric acid, the precipitate was analyzed. The data from the analysis were as follows:

0.1342 g of substance gave 0.0674 g Pt + Pd
0.1434 » » » » 0.1356 g AgCl

Calculated for $\|Pt2HxNH_3Py\|\ \|PdCl_4\|$	Found
% Pt + Pd . . . 49.83	49.86
% Cl 23.41	23.41

Trans-hydroxylamine-pyridine-diammonia-platinum chloride $|PtHxPy2NH_3|Cl_2$. 1 g of the non-electrolyte trans $|PtHxPyCl_2|$ finely ground is put into a porcelaine evaporating dish, 5 ml of water is added and then it is treated in the cold with 18% ammonia in slight excess with regard to the theoretical quantity, constantly being ground with a glass stirring rod or pestle. Under these conditions the non-electrolyte begins to dissolve noticeably, colouring the solution pale-green. The reaction goes on very slowly and is complete only after 1.5—2 hours. The instillation of ammonia into the inner sphere of the complex can be represented by the following equation:



$+ 2NH_3 \rightarrow |PtHxPy2NH_3|Cl_2.$

In this case heating harmfully affects the normal course of the reaction since $|PtHx3NH_3|Cl_2$ forms as a by-product which shows that pyridine

breaks away from the non-electrolyte $|PtHxPyCl_2|$. Strong heating causes the appearance of a brown amorphous precipitate which apparently consists of hydrolysis products.

An aqueous solution of the assumed tetramine $|PtHxPy2NH_3|Cl_2$ is evaporated in a vacuum exsiccator over sulphuric acid to small volume, 2—3 ml, for 2—3 days and then is treated with a mixture of alcohol and water. The precipitate which falls out is crystalline, in the form of long needles, and slightly greenish in colour. The substance dissolved well in water, dilute alcohol, not so well in concentrated alcohol. After being dried over sulphuric acid or by means of an air-dryer, the tetramine was analyzed and gave the following results:

0.1061 g of substance gave 0.0505 g Pt
0.1056 » » » » 0.0502 » »
0.1194 » » » » 0.0818 g AgCl
0.1018 » » » » 0.0697 » »

Calculated for $\|PtHxPy2NH_3\|Cl_2$	Found
% Pt	47.35	47.60; 47.54
% Cl	17.20	16.95; 16.93

The molecular electroconductivity of the tetramine measured at 25°C showed that the tetramine decomposes into one complex two-charged ion $PtHxPy2NH_3|^{++}$ and two simple chlorine ions — 2Cl′:

v	500	1000	2000	4000
μ_v	224.0	230.0	237.0	241.0

Trans-chloroplatinite $|PtHxPy2NH_3|\,|PtCl_4|$. A concentrated solution of this tetramine, acidified with dilute hydrochloric acid, in reaction with $K_2|PtCl_4|$ gives long red-coloured needles of the chloroplatinite sought only after standing 10—12 hours. The latter recrystallizes out of water acidified with 1—2 drops of 10% HCl without decomposing noticeably. After being dried at 108°, this substance gave acceptable analysis results:

0.1208 g of substance gave 0.0647 g Pt
0.1129 » » » » 0.0651 » »
0.1313 » » » » 0.1110 g AgCl

Calculated for $\|PtHxPy2NH_3\|\,\|PtCl_4\|$	Found
% Pt	57.55	57.50; 57.76
% Cl	20.91	20.91

A crystallographic and optical study of the crystals was kindly made by G. B. Bocky in the Crystallography laboratory of the Institute of General and Inorganic Chemistry.

The crystals of trans-chloroplatinite $|PtHxPy2NH_3|\,|PtCl_4|$ are monoclinic and belong to the prismatic type of symmetry. The simple forms observed were: {100}, {010}, {001}, {111}. The elements of the crystal are: $a:b:c=1.087:1:1.115$; $\beta=90\,000$. Refraction coefficients: $N_g=1.776$; $N_m=1.752$; $N_p=1.698$; $2V=65°$.

Trans-chloropalladite $|PtHxPy2NH_3|\,|PdCl_4|$. If an aqueous solution of the tetramine acidified with acetic acid is poured into a freshly-prepared concentrated solution of $K_2|PdCl_4|$ acidified with the same acid (but not the other way round), there appears a goldish-brown crystalline precipitate, which proves to be under the microscope in the form of wide, longish tablets — the chloropalladite sought. This substance dissolves well in water, not so well in alcohol (1 : 1) and very poorly in concentrated alcohol.

Aqueous solutions of the chloropalladite noticeably decompose in time to the metal. After being dried in the exsiccator over sulphuric acid for

24 hours, the chloropalladite was analyzed. The data of the analysis were as follows:

0.0840 g of substance gave 0.0410 g Pt + Pd
0.0846 » » » » 0.0432 » Pt + Pd
0.0924 » » » » 0.0900 g AgCl
0.1438 » » » » 0.1424 » »

Calculated for $\|PtHxPy2NH_3\|\|PdCl_4\|$	Found
% Pt + Pd . . .	51.19	51.00; 51.06
% Cl	24.06	24.11; 23.98

Trans-chloroplatinite $\|PtHxNH_32Py\|\|PtCl_4\|$. This is produced through the action of $K_2\|PtCl_4\|$ in the cold on a solution of tetramine, slightly acidified by 10% HCl. The precipitate which appears is pink in colour and in the form of microscopic crystals. The yield is almost equal to the theoretical result. The chloroplatinite decomposes when recrystallized from water; it dissolves poorly in water and dilute alcohol. After being dried at 107°, the substance was analyzed:

0.1052 g of substance gave 0.0554 g Pt
0.1188 » » » » 0.0912 g AgCl

Calculated for $\|PtHxNH_32Py\|\|PtCl_4\|$	Found
% Pt	52.74	52.66
% Cl	19.15	18.99

Trans-chloropalladite $\|PtHxNH_32Py\|\|PdCl_4\|$. The same tetramine solution as in the previous case was treated with a freshly-prepared concentrated solution of $K_2\|PdCl_4\|$, acidified with dilute acetic acid. A precipitate of a goldish-brown colour fell out almost at once. This precipitate when examined under the microscope proved to be in the form of long needles. Dark-brown crystals of the same shape precipitate from a hydrochloric acid medium. The chloropalladite dissolves well in water, not so well in dilute alcohol and does not dissolve at all in concentrated alcohol.

Aqueous solutions of the chloropalladite decompose noticeably, especially in sunlight, to the metal. The analysis data are in full accord with the formula ascribed to it:

0.0878 g of substance gave 0.0410 g Pt + Pd
0.0450 » » » » 0.0208 » Pt + Pd
0.0970 » » » » 0.0860 g AgCl
0.0600 » » » » 0.0538 » »

Calculated for $\|PtHxNH_32Py\|\|PdCl_4\|$	Found
% Pt + Pd . . .	46.31	46.70; 46.22
% Cl	21.76	21.95; 22.16

Conclusions. The following mixed hydroxylamine platinum tetramines and their derivatives have been produced for the first time:

trans-$\|Pt2HxNH_3Py\|Cl_2$; $\|PtHxNH_32Py\|\|PtCl_4\|$;
trans-$\|PtHxPy2NH_3\|Cl_2$; $\|Pt2HxNH_3Py\|\|PdCl_4\|$;
$\|Pt2HxNH_3Py\|\|PtCl_4\|$; $\|PtHxPy2NH_3\|\|PdCl_4\|$;
$\|PtHxPy2NH_3\|\|PtCl_4\|$; $\|PtHxNH_32Py\|\|PdCl_4\|$.

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CHEMISTRY

REGULARITIES OF FORMATION AND STRUCTURE OF PROTECTIVE FILMS ON METALS

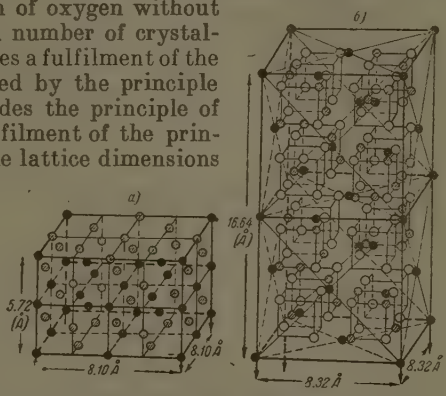
By P. D. DANKOV

(Communicated by Wl. A. Kistjakowsky, Member of the Academy, 8.IV.1939)

1. For some time past the problems of the structure and the mechanism of formation of films which cause metals to become passive have been subjected to animated discussion(1). At present we have sufficiently complete data as far as oxide films on iron are concerned. In connexion with this it becomes possible to establish a number of important regularities governing the formation and the structure of protective films.

The characteristic peculiarity of a passivating film on iron [γ-Fe_2O_3 and Fe_3O_4(1,2,3)] is its sufficient continuity and comparatively high adhesion to the basic metal. Together with this, optical(4) and electronographic (1,2,3) data point to the extremely small thickness of the film which ranges from 10 to 30 Å (in special cases the film thickness may be larger). Taking into account the insufficient accuracy in the measurements, one may assume that the film thickness is determined by the imposition of some two-three elementary cells of the cubic oxide (γ-Fe_2O_3 or Fe_3O_4), whose linear dimensions are close to the value 8.32 Å (or 8.38 Å). It follows from this that the oxidation process of a metal under the conditions of room temperature is extremely close to the phenomena of the chemical adsorption of oxygen, a discussion of which was given by De-Boer and his co-workers (5). The material cited below confirms this point of view. Evidently, the diffusion (in the usual sense of this word) transitions of oxygen into the crystalline lattice of iron do not take place during low temperature oxidation (e. g., at room temperature), since in order to form an oxide film $\sim$ 16 Å in thickness (which corresponds to the doubled height of the elementary γ-Fe_2O_3 or Fe_3O_4 cell), it is sufficient for oxygen to penetrate only into two upper surface layers of the elementary iron cells. Fig. 1 elucidates this point. Fig. 1,*a* shows two layers of elementary cells of α-iron belonging to one of the monocrystals of the metallic surface. The crystal is orientated with the [100] direction perpendicular to the surface of the sample. Oxygen molecules or molecules of some other oxidizing agent (e. g., K_2CrO_4, $K_2Cr_2O_7$, HNO_3), which arrive either from the gas phase or the liquid phase, are directly adsorbed on the crystal surface. Passing over the small potential hill of the potential energy curve(5), the system metal-oxidant gives a combination of the atoms of 16 elementary iron cells, of which 8 are whole and 16 are halves (the cells are divided in half by the diagonal planes), and of the number of oxygen atoms, which corresponds to two adjacent elementary cells of γ-Fe_2O_3 and Fe_3O_4, constructed according to the spinel lattice type. Both oxide cells lie upon each other. Thus, oxygen atoms do not have to penetrate deeply into the iron; on the contrary, some iron atoms stretch closer to the surface. Oxygen atoms filling up the iron cells force the latter to swell. At the orientation of the microcrystals pointed out, the swelling proceeds in one direction—upward (in the [100] direction). The

construction of the oxide cells is carried out in accordance with preseng concepts of a lattice of the spinel type and in agreement with the data of Dankov and Shishakov (1) and also of Nelson (2) on the application of the principle of orientation correspondence between the lattices of γ-Fe_2O_3 (or Fe_3O_4) and of the iron undergoing oxidation; in the given case the [111] direction in the iron lattice is practically parallel to the [$\sqrt{2}$ 10] direction in the lattices of γ-Fe_2O_3 and Fe_3O_4 which indicates a regular swelling of the iron lattice during the absorption of oxygen without any change in the orientation of a number of crystallographic directions, i. e., it indicates a fulfilment of the correspondence which is determined by the principle of orientation correspondence. Besides the principle of orientation correspondence, the fulfilment of the principle of the correspondence of the lattice dimensions of the metal and the film formed happens to take place and to be necessary for the protective films. Indeed, upon the indicated orientation of the metal and oxide lattices, the doubled diagonal of the cubic plane of the iron lattice coincides with the edge of the oxide lattice; this coincidence is accurate to 2% (8.32 Å—2·4.05 Å=0.22 Å). The difference may be compensated by the distortion of the oxide and metal lattices which has already been observed in the case of iron. The presence of such a correspondence of dimensions causes the adhesion of the protective film to the metal, because only under these conditions the maximum binding of the film to the metal by way of common iron atoms is preserved.



Scheme of transition of crystalline lattice of α-iron into that of Fe_3O_4 and γ-Fe_2O_3: a) part of α-iron crystal limited by 4 planes (110) of dodecahedron (lateral) and by 2 cubic planes (100) (upper and lower); б) 2 cells of Fe_3O_4 and γ-Fe_2O_3, arising at interaction of O with a part of the Fe-crystal shown in a) · (● and ◍—Fe-atoms, ○—O-atom).

In the general case various orientations of microcrystals occur on the surface of a sample. As a more detailed analysis of cases differing from the one discussed above shows (e.g., orientation of the [110] or the [111] direction perpendicular to the surface), the oxidation process in this case occurs under less advantageous conditions, since upon the fulfilment of the principle of orientation correspondence*, the swelling of the metal lattice should proceed along the surface. This will be impossible if the crystals are sufficiently densely distributed on the surface, or will be retarded owing to the influence of cohesive forces which must be overcome in the development of the oxidation process. In the case of the development of the oxidation process, one should here expect a decrease in the adhesion of the film to the metal. The process will be retarded also owing to the fact that oxygen must penetrate to a considerably greater depth, which is connected with extreme difficulties (diffusion at room temperature). One may also expect that in these cases an oxide film is not formed at all, and the process stops at the stage of chemical adsorption without the formation of a crystalline oxide lattice. Certainly, in the case of very small crystals (combinations of some 10—20 elementary cells) of iron the circumstances mentioned will not preclude

* A comparison of the papers already cited (1) and (2) with the work of Mehl (6) indicates the applicability of this principle to the cases of various orientations of iron crystals.

oxidation, since practically the whole ultra-microcrystal will be transformed into the oxide. The fact that in our experiments[1] and in Nelson's[2] experiments it was established that iron is oxidized with its crystals orientated with their [111] planes parallel to the surface may be easily explained by that very circumstance that the dimensions of the original iron crystals were very minute, and only their projections (peaks, ridges), which easily oxidize completely, were subjected to X-raying (by the „reflexion"method).

The presence of natural cracks on the surface of small single crystals (mosaic structure) will also be an extremely important factor during oxidation. In connexion with the established dependence of the degree of oxidation upon the dispersity, one should note that a polished surface which, according to a number of investigators, consists of amorphous metal should easily react with oxygen and in so doing should yield an amorphous or an extremely dispersed oxide; this is in accord with some theses advanced by Kistjakowsky[7].

2. The considerations developed with respect to the process of the formation of a passivating oxide film on pure iron may be enlarged so as to include a wide variety of metals and passivating films of various types.

Stainless steels, which easily renew the protective oxide layer on their surfaces, are of particular interest. Acccording to Evans[8] chromium oxide is present in the composition of the passivating oxide film formed on stainless steels; in other words, the possibility that compound protective films may exist is justly recognized. Compound films can be conceived also in the case of alloyed steels and in the formation of artificial protective coverings on iron (calorizing, sherardizing, chromium plating, and other methods of formation on the surface of iron alloys), made of other metals and their alloys with iron. The formation of a complex film presupposes definite proportions between its components. In the case of stainless steel these components are oxides of iron and chromium. A question arises whether they, taken either separately or in some combination, will satisfy the above formulated principles of orientation correspondence and dimensional correspondence. It is as yet insufficiently clear how chromium oxide taken separately behaves in this respect, but the combination of ferrous oxide and chromium oxide may give the correct proportions, which follows from the comparison of the lattices of γ-Fe_2O_3 (or Fe_3O_4) and $FeCr_3O_4$ given below. It happens that all these lattices belong to the same type, and what is still more remarkable is that the constants of these cubic lattices almost coincide, as is seen from the table.

Substance	Lattice dimension	Substance	Lattice dimension
$NiAl_2O_4$	8.04	$MnFe_2O_4$	8.46
$MgAl_2O_4$	8.07	$MnCr_2O_4$	8.49
$ZnAl_2O_4$	8.10	$TiFe_2O_4$	8.50
$EeAl_2O_4$	8.12	$CuAl_2O_4$	8.07
$MnAl_2O_4$	8.26	$CoAl_2O_4$	—
$FeCr_2O_4$	8.35	λ-Fe_2O_3	8.32
$FeFe_2O_4$ (Fe_3O_4)	8.38	λ-Al_2O_3	7.90
$ZnFe_2O_4$	8.42	λ-Cr_2O_3	—

It is seen from the table that the group of substances presented is remarkable in the respect that its representatives, being composed of the most diverse combinations of seven atoms (4 oxygen atoms, 2 trivalent atoms, and 1 bivalent atom) and preserving a structure similar to the spinel type, differ quite insignificantly in the values of their lattice constants. Of the substances presented in the table, the compound $FeCr_2O_4$ is the closest to Fe_3O_4 with respect to the value of its lattice constant. Therefore the pro-

tective film on stainless steel, corresponding in its composition to $FeCr_2O_4$, will satisfy sufficiently well the principles of orientation and dimensional correspondences. One may expect the same in the case when pure iron is made passive in solutions of chromates. Evans([8]) also points to this possibility. In the last two cases one may expect the formation of lattices belonging to the γ-Fe_2O_3 type (instead of to the Fe_2O_4 type) for the compound film also (e. g., $Fe_2O_3 \cdot Cr_2O_3$); such a crystalline structure has not been mentioned in the literature as yet. However, similar to γ-Fe_2O_3 which is a product of the careful oxidation of magnetite (Fe_3O_4) one may expect the formation of a lattice consisting of $Fe_2O_3 \cdot Cr_2O_3$ which will also preserve the form typical for spinel lattices. Here one may expect a decrease in the lattice constant which will lead to a greater precision in the coincidence of the dimensions of the metal and oxide lattices.

It is not difficult to understand that the substances presented in the table can be formed on the surface of iron coated with the aid of one of the mentioned methods of depositing artificial protective films. Evidently, the compounds $FeAl_2O_4$, $ZnFe_2O_4$, $MnFe_2O_4$, $TiFe_2O_4$, $FeCr_2O_4$ may form a protective film on a metal and in so doing the principles of the orientation and dimensional correspondences will hold here also.

3. Passing to the problem of the protection of other metals, it is necessary, even though briefly, to dwell upon aluminum and chromium which, as is known, easily form a protective oxide film on their surfaces. As Archarov([9]) correctly noted, the passivating films on these metals possess a crystalline lattice of the same type, namely a spinel type; we emphasized this by including data for these oxides in the table. An analysis of the problem shows that in this case also the required relations between the metal and the oxide lattices are present, and this is the reason for the high resistance to corrosion in aluminium and chromium. In contrast to iron, the orientation of the lattice planes of the oxide is the same as the orientation of the corresponding metal planes. Additional experiments are necessary, of course, in order to check this.

It should be noted, furthermore, that aluminium alloys of various kinds with other metals (Mg, Zn, Ni, Fe, and others) may (and sometimes actually do) possess a high stability also, because to these alloys there correspond oxides forming lattices of the spinel type (table). For them and for the lattices of the alloys one may easily find dimensions which satisfy both principles of correspondence. Among the other metals which form comparatively stable protective oxide films on their surfaces, it is of interest to mention zinc and nickel. For the former of these metals one obtains a coincidence of dimensions and orientation for the metallic and oxide lattices which is in agreement with Finch's data([10]). For the latter one, it happened to be difficult to find the correct relations. Owing to the fact that the tabulated data for nickel and its oxide are contradictory, we shall leave the analysis of this problem open.

4. The study of protective films, differing from oxide films, corroborates the possibility of applying to them the principles of orientation and dimensional correspondences. As examples one may mention the protective film of lead sulfate on lead (an example of a complex correspondence), iron nitride on iron (also a complex correspondence), and a number of other cases, the analysis of which will be presented in a more detailed paper.

5. The examples of the applicability of the regularities discussed to cases of technical protective films on metals are a good criterion of the use that can be made of the theoretical scheme of the formation and structure of surface films that has been developed in this paper. However, these regularities refer not only to the cases of protective films on metals. Various

low temperature (i. e., without the interference of diffusion) phase transformations developing on the surface of a solid must satisfy the principles of orientation and dimensional correspondences between the original crystal lattice and the crystal lattice formed.

6. The new ideas developed in the present preliminary communication may be briefly formulated as follows. 1) The processes of the origin of protective films on metals are analogous to the phenomena of the formation of surface oxides([5]), because the transformation develops only in the first layer of the elementary cells of a metal.

2) The formation of two layers of elementary cells of iron oxide (16—17 Å) occurs as the result of unidirectional „swelling" of the elementary surface cells of iron, and there is an indication of the possible presence both of γ-Fe_2O_3 (in the upper layer) and of Fe_3O_4 (in the lower layer).

3) On the basis of the data for oxide films on iron there are formulated the principles of orientation correspondence and correspondence of dimensions of the conjugate crystalline lattices of the metal and oxide. The applicability of these principles to the given system metal-oxide is a criterion of their adhesion and protective properties.

4) An analysis of the problem concerning the various orientations of microcrystals on a metal surface leads to the possibility of the existence of some portions of the metal which are covered not with the crystallized oxide, but with a layer of chemically adsorbed oxygen.

5) An analysis of the problem of the dispersity and mosaic structure of metallic microcrystals indicates the possibility of the formation of a highly dispersed (up to an amorphous state) layer of oxides on the polished surface.

6) Upon a comparison of the data for protective layers on stainless steels and on metal coatings (calorizing, sherardizing, chromium plating, etc.) with the structural data for compounds of the spinel type (table), the applicability of the principles of orientation and dimensional correspondences to complex (composite) protective oxide films becomes apparent.

7) The applicability of the principles pointed out can be extended to cover the cases of protective oxide films on aluminium, chromium, zinc, and other metals, and on various kinds of alloys also.

8) Using as examples protective films which differ from oxide films (e. g., $PbSO_4$, Fe_2N, FeSi, etc.), there is disclosed the applicability of the correspondence principles to these cases also.

9) A method is suggested to improve the properties of protective films by means of an artificial orientation of the metallic crystals before the formation of the protective layer and to form these films.

10) There is pointed out the possibility of a general applicability of correspondence principles to cases of phase transformations (upon the formation of a new solid surface phase) on the surface of a solid.

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GEOLOGY

ON SOME SYENITE ROCKS OF THE SUPSA RIVER (WEST GEORGIA)

By G. G. KAZAKASHVILI and P. A. TOPURIA

(Communicated by A. F. Fersman, Member of the Academy, 5.IV.1939)

Along the Supsa river in West Georgia (Fig. 1), two peculiar young dike intrusions of a syenite composition have been encountered by the authors. The peculiarity of the first intrusion, exposed 15 km upstream of the village of Tav-Surebi, consists in its differentiation—a different composition of rocks at the lower and upper portions of the dike; the peculiarity of the second one, detected at the middle part of the Supsa river near the Didi-Vani settlement, consists in the petrographic composition of the composing rock. The latter has been named by the authors analcime syenite, since it is a transitional rock from typical syenites to teschenites, widely known in Georgia. The Didi-Vani intrusive is accompanied by veins of analcime gabbro-syenite and trachyte, the former being obviously younger than the main intrusion, since one of these veins is cutting the analcime syenite intrusion.

Both these intrusions occur among Middle Eocene beds of andesite breccia and flysch („porphyritic Eocene") in the form of dikes, producing in them contact alterations, which are especially intensive around the syenite intrusion upstream of the village of Tav-Surebi. The above said points to the post-Middle Eocene age of the intrusions; by analogy with other Georgian intrusions, some relation is probable to exist between the Supsa rocks and the Priabonian phase of the Alpine folding.

1. Syenite Intrusion 15 km upstream of the Village of Tav-Surebi. Outcrops of the intrusion along the bed of the Supsa river are exposed at a distance of about 1.5 km. Both contacts are dipping N-NW. The impression made is that of a dike-like body of about 500—600 m in thickness; the upper and lowermost outcrops along the river should be regarded as the roof and foot of this dike. The difference in composition may be well seen when quantitatively comparing the mineralogical composition of specimens collected successively from the foot to the roof (Table 1).

A maximum thickness is attained by the middle syenite part stretching along the bed of the river over a distance of about 500 m. The syenite diorites of the lower portion of the dike are 300 m thick. The upper syenite porphyres (syenite aplites)—about 200 m. The transition from the former to the latter rocks is quite gradual.

The increase in the acidity of the rock (Table 2) proceeds parallel with the decrease in the total specific gravity of the whole rock mass from the foot to the roof. Moreover, the rocks of the roof differ from those described above also by their structure: the syenite and syenite-diorite rocks of the base are hypidiomorphous granular structures the syenites aplite of the roof—porphyritic.

Table 1

Quantitative Mineralogical Composition of the Rocks of the Supsa Syenite

Mineral constituents	Lower part of the intrusion		Middle part		Upper part
	Syenite-diorites, %		Syenites, %		Syenite-porphyres
	№ 31	№ 36	№ 37	№ 38	№ 40
Quartz	—	—	0.2	2.3	4.8
Anorthoclase	17.6	35.3	75.4	83.7	91.6
Plagioclase	55.7	43.6	9.5	7.8	1.9
Nos plagiocl.	35—43		18—23		8 (less freq. 20)
Biotite	10.7	9.1	3.2	0.5	
Augite	13.2	8.2	8.2	4.6	1.6
Magnetite	2.8	3.8	3.5	1.4	
Total	100.0	100.0	100.0	100.3	99.9
Spec. gravity:					
Measured	2799	—	2697	2678	—
Calculated	2750	2705	2690	2654	—

Table 2

Chemical Analyses of Rocks and Minerals of the Supsa Intrusions

	1	2	3	4	5	Magmatic formulae according to Loewinson-Lessing
						—
SiO_2	53.47	58.30	59.88	63.94	63.22	1) $1.41\ RO.R_2O_3 \cdot 7.98\ SiO_2$
TiO_2	0.64	0.39	0.53	traces	0.02	$R_2O : RO = i : 5.1$
Al_2O_3	18.31	17.34	19.59	20.02	19.52	$\alpha = 1.8$
Fe_2O_3	7.11	4.74	3.05	0.40	0.47	—
FeO	1.78	0.78	1.46	—	0.25	2) $1.54\ RO.R_2O_3 \cdot 4.94\ SiO_2$
MnO	0.22	0.14	0.18	—	traces	$R_2O : RO = 1 : 1.58$
CaO	7.79	5.22	2.75	0.58	0.96	$\alpha = 2.16$
MgO	4.18	3.37	2.10	0.07	traces	
BaO	—	—	—	0.17	—	—
Na_2O	2.65	5.17	4.51	3.60	5.25	3) $1.0\ RO.R_2O_3 \cdot 4.6\ SiO_2$
K_2O	1.72	3.81	4.15	10.86	8.70	$R_2O : SO = 1 : 1.02$
SO_3	0.11	0.23	0.11	—	—	$\alpha = 2.5$
P_2O_5	0.25	0.04	traces	—	—	
H_2O+	0.54	0.26	0.73	—	—	
Loss on ignit.	1.48	0.32	1.28	0.60	—	
Total	100.45	100.11	100.32	100.24	99.39	—

1) Syenite-diorite of the lower part of the syenite intrusion. 2) Syenite of the middle part of the intrusion. 3) Analcime syenite. 4) Anorthoclase from the syenite intrusive. 5) Anorthoclase from the analcime syenite.

Analyser: № 1, 2, 3,—R. Nakashidse; № 4—V. A. Moleva; № 5—P. Volkov.

The character of individual minerals of the rock is uniform throughout the intrusion. The composition of the plagioclase alone is variable as shown in Table 1. The anorthoclase ($2V = -60°$), as is always the case in

neointrusions, is essentially potassic and contains about 30—35% of albite, so far as it may be judged from its refraction of light (N_g=1.528±0.002 and N_p=1.521±0.002). Aside from such chemically bound albite, it shows also a slight perthitization in the portions lying nearest to the roof. The chemical analysis of anorthoclase is given in Table 2, column 4. A recalculation of these data gives Or—64.5%, Ab № 8—35.3%, Ce—0.2%.

Dark-coloured minerals: monoclinic pyroxene of the augite series (CN_g= 38—42°; 2 V=+45+50°) and biotite. Hornblende, secondary; at the lower portions of the intrusion it partially replaces the monoclinic pyroxene, and near the roof the replacement is almost complete. Chlorite is developed at the expense of the two primary minerals. Accessories: apatite, zircon and magnetite.

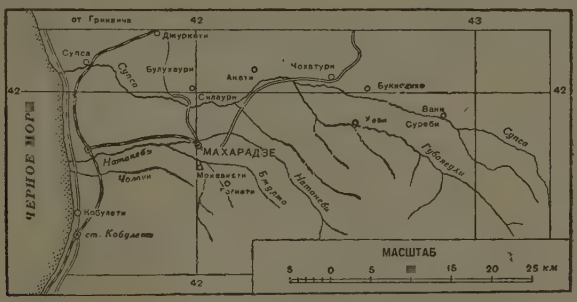

Fig. 1.

At the contacts of the intrusion with rocks of the porphyritic Eocene are formed anorthoclase-oligoclase hornfels with biotite (anorthoclase—39.7%, oligoclase № 13—39.4%, biotite—18% and quartz—1.8%) analogous to similar rocks of the Vakis-Dzhwar intrusion(1).

2. Intrusion of Analcime Syenites near the Village of Didi-Vani. As shown in Fig. 2, the intrusion is confined to a small fault at the northern limb of the overturned anticline of the flysch, having an approximately latitudinal strike. The intrusion occurs here also as a dike with a SO—O dip of the contacts. The thickness of the intrusive is about 500—600 m. In the surrounding rocks numerous aplitic apophyses are observed.

The Vani intrusive is more or less homogeneous. Its bulk is composed of a idiomorphous granular rock of a pinkish-grey colour; the marginal parts are more fine-grained and somewhat darker, the same as the apophyses. The quantitative proportions of the chief minerals composing the typical rock are shown in Table 3.

Table 3

Minerals	Volume in %
Anorthoclase	67.1
Plagioclase № 47	16.3
Analcime	10.3
Dark minerals	6.4

By their character these minerals are essentially different from those of the Tav-Surebi syenite. The anorthoclase possesses a considerably lesser axial angle (average for $2V$=—43°; single determinations show values from—40° to—48°) and a much higher content of the albite particle which tells on its refraction of light (N_{g1}=1.531±0.002), (N_{p1}=1.524±0.002) and may also be clearly seen from the analysis of selected material (Table

2, column 5) a recalculation of which shows: Or=49.59 and Ab № 9=50.44; this fully concurs with the optics. The anorthoclase forms idiomorphic crystals, pelitized and replaced by natrolite.

The plagioclase forming large crystals is usually completely replaced by zeolites, analcime and natrolite; sometimes it is bordered with anorthoclase. In the rare preserved areas the authors succeeded in establishing its andesitic nature (№ 47—45). The dark minerals are represented by predominating reddish-brown biotite, monoclinic pyroxene (average $CN_g=39°$, $2V=+53°$) and ordinary hornblende. Aside from the replacement of feldspars the analcime forms also independent segregations in the angular spaces between

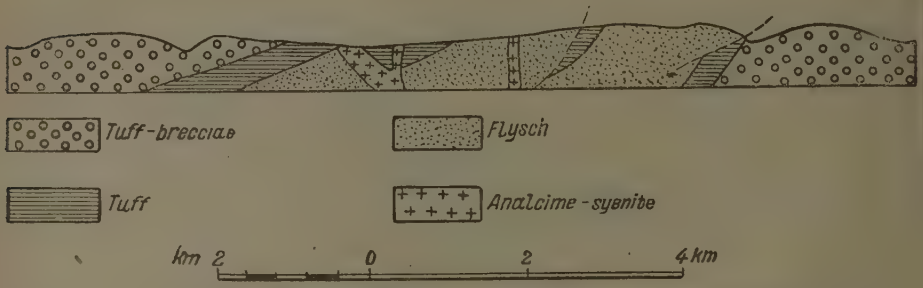

Fig. 2. Section along the Supsa river between the villages of Bukis-Zikhe and Didi-Vani.

the other constituents of the rock. Inclusions of idiomorphic crystals of plagioclase, dark minerals and needles of apatite are often encountered in analcime. Occasionally the apatite forms elongated needles up to 0.5 mm in length. Magnetite is present.

It has been previously observed that the intrusion is accompanied by veins of analcime trachyte and analcime gabbro-syenite. The former is distinguished from analcime syenite of the main intrusive by its porphyritic structure and the trachytic habit of the ground mass composed of microlites of anorthoclase, minute grains of magnetite and a quite negligible quantity of a brownish dull base. The phenocrysts are represented by anorthoclase ($2V=-56°$), partly by analcimized albite № 9—10 ($N_g=1.538$) and biotite. The second, the gabbro-syenite, is characterized by a predominance of plagioclase in the feldspar portion of the rock and by its highly basic bytownitic composition (№ 81—90). A large quantity of monoclinic pyroxene ($CN_g=35—41°$; $2V=+45,+52°$). The anorthoclase is fresh and forms borders around the plagioclase. The quantity of biotite is small. The quantity of analcite and natrolite is less than in the analcite syenites.

Conclusions. From the two neointrusions described the most interesting is the second, a differentiated intrusive of analcime syenite in the neighbourhood of the village of Didi-Vani. The composing rock is a connecting link between the normal syenites of the Supsa river and other places of Transcaucasia, on the one hand, and other analcime-containing basic rocks, on the other.

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PETROGRAPHY

ESSAY OF A RATIONAL QUANTITATIVE MINERALOGICAL CLASSIFICATION OF GRANITOIDS

By B. M. KUPLETSKY and O. I. KOVALEVA

(Communicated by F. J. Loewinson-Lessing, Member of the Academy, 31.III.1939)

During the last years a series of works by Niggli, Traeger, Johansen, Andreatta etc. appeared in the petrographic literature, in which attempts were made to systematize the accumulated material on the quantitative mineralogical composition of rocks, and to construct a classification of rocks on the basis of their real and not normative mineralogical composition, which has been used by American petrographers in their well-known but purely conventional and artificial classification. Considering the necessity of giving a natural classification of rocks, one of the authors began to systematize as early as in 1932 the material available on the mode quantitative mineralogical composition of igneous rocks, both from literary data and from numerous written records and other materials at disposal of the Geological Institute of the Academy of Sciences.

After a proper treatment the material accumulated in this way made it possible to work out essay of a rational classification of granitoids now offered to the reader. This group of rocks comprises a number of transitional rock types from granites to syenites and diorites; these rocks are named differently by different authors and that is why some rocks similar or analogous as to their mineralogical composition are mentioned in literature under four or five different names. In consequence of this the geologists speak different languages, and it is almost impossible to compare the descriptions made by individual geologists. Now this badly affects mining practice, since the numerous deposits of rare metals are associated with different types of granitoids and therefore a precise nomenclature and correct characteristics of the rocks are here of particular importance. The present work which is an attempt to introduce more clearness into the petrographic terminology and to establish a rational nomenclature, is based upon 1700 modes of granitoids, compiled from literature and the stocks of the Geological Institute of the Academy of Sciences.

Of these 1100 analyses refer to direct calculations of the quantitative mineralogical composition of the rocks made under the microscope and have been worked up by B. M. Kupletsky, and about 600 compositions have been obtained by means of recalculating chemical analyses to the mode quantitative mineralogical composition and have been worked up by O. I. Kovaleva under the direction of B. M. Kupletsky. The authors undertook the task of devising a working diagram, which would allow the granitoids to be determined quickly, proceeding from their quantitative mineralogical composition. Since the granitoids are composed, as a rule, of four components:

quartz-orthoclase—plagioclase—coloured and secondary minerals, we have to do here with a polycomponent system which may be represented by a spatial diagram—a tetrahedron, in whose vertexes the above minerals are placed. To pass from a spatial representation to a planar one, it is necessary to deal with several sections of the tetrahedron which would be determined by a certain relation of two components. Then the spatial diagram will be reduced to several triangular sections, giving the representation of the system in a plane.

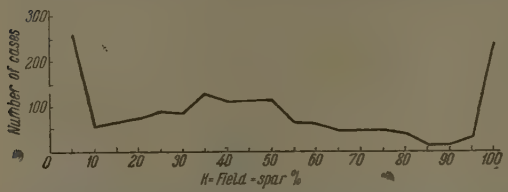

Fig. 1. Cumulative diagram of distribution of granitoids according to K-feldspar content in the total of feldspars of the rock (1720 specimens).

As chief co-ordinate for the selection of the tetrahedron sections the ratio potassium feldspar—total sum of feldspar in the rock has been taken, and then the following minerals have been placed in the apexes of each triangular section: quartz (Qu), feldspar (Fel), coloured and secondary minerals (Fem).

In order to avoid an accidental selection of sections of the tetrahedron, a curve has been plotted of the distribution of the granitoids according to their K-feldspar content in the totality of the feldspars of the rock (Fig. 1). The K-feldspar is accompanied also by albite up to the 10th №, because, although albite belongs to plagioclases, it is at the same time a typical alkaline feldspar, and some petrographers (Niggli, Traeger etc.) are classing albite among the alkaline feldspars. The alternation of maximum and minimum marked in this curve allowed the following most rational sections of the tetrahedron to be chosen.

Section I includes rocks with 0—10% K-feldspar to 100—90% plagioclase. The section embraces the family of diorites, tonalites, plagioclase granites and trondjemites. Section II comprises rocks with 10—30% K-feldspar to 90—70% plagioclase. These rocks embrace a part of diorites and granodiorites. In section III enter the rocks with 30—60% K-feldspar to 70—40% plagioclase. To these are referred monzonites, quartz monzonites and adamellites and the basic group of granites. In section IV rocks are distributed containing 60—80% K-feldspar to 40—20% plagioclase and referring to the group of syenites, shonkinites and granites. Lastly, in section V are included syenites, alkaline and alaskite granites containing 80—100% K-feldspar to 20—0% plagioclase.

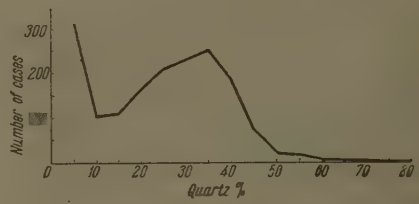

Fig. 2. Distribution curves of granitoids according to their quartz content (1720 specimens).

Taking as the main co-ordinate of granitoid division the ratio of K-feldspar to plagioclase in the rocks and not the relation of the anorthite molecule to the orthoclase and the albite molecules, as done by Shand and Andreatta, the authors wished to avoid superfluous artificial recalculations and give their classification the simplest and most natural aspect.

Besides it is not always possible to determine the composition of plagioclases in granitoids with due precision, especially in zonal plagioclases, and, lastly, the composition of plagioclases in granitoids usually varies over a comparatively narrow range from oligoclase to andesine, and there-

fore the number of plagioclase is of no importance in granitoids except the case of monzonites.

When separating monzonites from syenite-diorites, it is correct to refer to monzonites only such rocks of sections II and III of our tetrahedron, in which the plagioclase is more basic than andesine No 40 and where the idiomorphism of the plagioclase determines the monzonite structure of the rock.

All the granitoids have been placed in corresponding sections of the tetrahedron according to their quantitative mineralogical composition, and thus it has been possible to outline in every section the fields of development of igneous rocks of the granitoid groups.

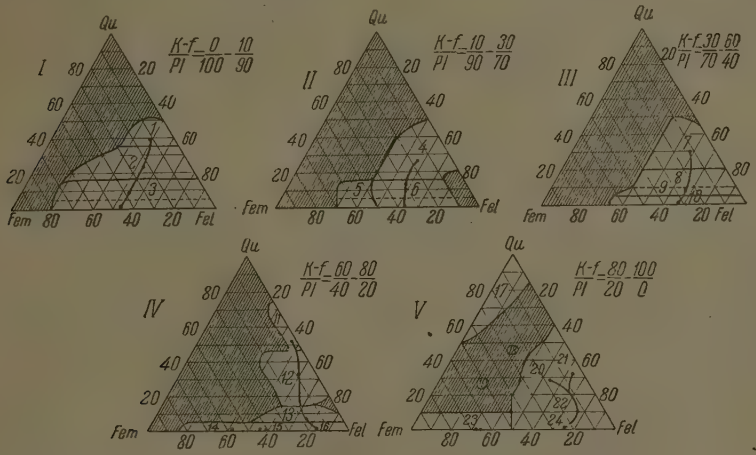

Fig. 3. *1*—Plagiogranites and trondjemites. *2*—Tonalites and granodiorites. *3*—Diorites and quartz-diorites. *4*—Granodiorites. *5*—Melodiorites. *6*—Diorites and monzonites. *7*—Granites. *8*—*9*—Addamellites and quartz monzonites. *10*—Monzonites and syenito-diorites. *11*—Leucogranites. *12*—Granites. *13*—Quartz syenite. *14*—Shonkinites. *15*—Monzonites. *16*—Syenites. *17*—Greisens. *18*—Moyite. *19*—Rockallite. *20*—Alkaline granites. *21*—Alaskites. *22*—Quartz syenites and granosyenites. *23*—Shonkinites. *24*—Syenites.

In every section of the tetrahedron further subdivision of rocks was made according to the quartz content in rocks. The statistical summarized curve of frequency of occurrence of granitoids with a given quartz content (Fig. 2) enables such main groupings to be established in every triangular section of the tetrahedron which have somewhat variable boundaries in individual sections:

Rocks with 0— 5% quartz—syenites, diorites and monzonites
» » 5—10% » —analogous quartz rocks
» » 15—20% » —transitional group of rocks (granodiorites, adamellites, granosyenites)
» » 25—40% » —basic group of granites and granodiorites
» » 45% and above—rocks highly oversaturated with silicic acid, leucogranites, greisens etc.

As pointed out above, the authors have independently of each other constructed two series of diagrams: one, based upon the mode of the rocks, obtained directly by calculations under the microscope and based upon 1100 data (Kupletsky), and the other—based upon modes calculated from chemical analyses, which included 600 compositions of granitoids (Kovaleva). The present work is accompanied by a cumulative diagram (Fig. 3)

obtained from microscopic counts of the modes of granitoids as being based on more extensive material.

Both diagrams constructed on the basis of two different materials essentially concur with each other, and certain differences in the limitation of the fields of stability between individual granitoid families have been observed only in sections I and V.

In section I, in the diagram accompanying the present work, to the plagiogranites are referred rocks containing more than 35% quartz, while to tonalites are referred rocks with 15—35% quartz. In the diagram, constructed by Kovaleva from the structures calculated from chemical analyses, the boundaries of the tonalite field are limited by the 10—30% quartz content and correspondingly to plagiogranites are referred rocks containing over 30% quartz.

In section V the principal difference of the diagrams based on different materials consists in that the field of alkaline granites is beginning in the published diagram with 20% quartz, while in the materials given by Kovaleva the boundary between granosyenites and alkaline granites lies at 30% quartz, according to the compositions calculated.

These differences show that a part of the boundaries of stability fields of individual granitoid families shown in the diagrams are rather conventional; besides, this follows from the course of the curve in Fig. 2, in which obvious breaks are absent. Nevertheless, the main types of granitoid families may be outlined at present with sufficient precision.

The cumulative diagram given in Fig. 3 shows 5 sections of the tetrahedron, in which the fields of stability of individual granitoid families are isolated. This diagram allows the granitoids to be classified directly according to their quantitative mineralogical composition obtained by counts under the microscope. After having calculated under the microscope the mineralogical composition of any granitoid and counted up the ratio of K-feldspar in the total amount of feldspars of the rock, we can determine directly the section, to which our rock is to be referred. Having plotted the point of the rock in the corresponding triangle, we determine the family, to which the given rock belongs. If for instance we have a rock composed of 25% quartz, 40% oligoclase, 30% K-feldspar and 5% coloured minerals, the ratio of K-feldspar to plagioclase in the feldspar portion of such a rock is 43 : 57. The rock is referred to section III of the tetrahedron where, according to its coefficients 25 quartz—70 feldspar—5 coloured minerals, it is placed into the field of development of granites which determines the name of the rock.

Among the different sections of the tetrahedron the granitoids are arranged not quite uniformly, e. g. section IV comprises so few rocks that here the fields of development of individual families were found to be restricted, and for the mineralogical compositions obtained by recalculation from the chemical composition of granitoids it proved impossible to isolate fields for individual granitoid families in this section owing to the lack of material.

Perhaps later on, when more material is accumulated, it will be necessary to reduce this section, and to distribute the rocks belonging to it between sections III and V.

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Institute of Geological Sciences.

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CRYSTAL CHEMISTRY

THE COURSE OF ORIENTATED DISPLACEMENT OF MAGNETITE BY IRON

By V. A. FRANK-KAMENECKIJ

(Communicated by A. E. Fersman, Member of the Academy, 2.IV.1939)

In one of the recent numbers of C. R. Acad. Sci. URSS (1) a paper appeared written by N. Buynov, M. Zhuravlev, A, Komar and G. Chufarov which deals with the orientation of iron crystals, occurring when magnetite is reduced with hydrogen. These investigators made use of a monocrystal of magnetite; after reduction at 500° C, there was formed on it a monocrystalline layer of metallic iron several thousandths of a millimeter in thickness. As an X-ray study showed (rotation and Laue methods), the orientation of this iron layer on the magnetite was such that its [001] axis was parallel to the [001] axis of the magnetite. Katzaurov (2) also pointed out the presence of a definite orientation of iron upon the reduction of magnetite.

During the past few years a number of investigations have appeared which make use of X-ray and electronographic methods for a study of the orientation of crystals formed during the transformations of solid phases (displacements).

It was established that while the films of ZnO upon Zn (3), FeO upon Fe (4) and Cu_2O on Cu (4,5) have a fixed orientation relationship, the films of Al_2O_3 upon Al (6) and WO_2 upon W (7) are oriented arbitrarily. Mehl and McCandless (8) established that, upon the oxidation of wüstite (FeO), a fixed orientated film of magnetite (Fe_3O_4) is formed on it. The regular interpenetrations of magnetite and haematite have already been known (9) for some time past.

Numerous investigations [(10–14) and many others] have shown that the mutual orientation of regularly interpenetrating crystals in most of the cases is caused by the structure of the interpenetrating nets (similarity of the pattern and proximity of the values of the parameters of their multiples). Royer (11) proposed such interpenetrations to be called epithaxy. Mehl, McCandless and Rhines in an analogous manner explained the formation of wüstite on iron (4) and magnetite on wüstite (8).

In the present communication an explanation is given for the fixed displacement of magnetite by iron described in the previously mentioned papers (1,2).

X-ray data for the structures of magnetite (15,16) and α-iron (17,18) permit of an examination of the structures of those planes parallel to which the displacement of magnetite by iron occurs. The two-dimensional (001) nets of magnetite form a pattern having the configuration of a centered square. If we omit the $O^{\cdots}$ ions, which in magnetite form the almost ideal, densest

cubic packing, then the $Fe^{\cdot\cdot}$ ions (large circles) and the $Fe^{\cdot\cdot\cdot}$ ions (small circles) will be distributed as shown in Fig. 1. Here the $Fe^{\cdot\cdot}$ ions possess a quadruple co-ordination, while the $Fe^{\cdot\cdot\cdot}$ ions a sextuple; accordingly we will examine individually the layers formed by these various ions. An elementary magnetite cell has a side $a=8.42$ Å. In the magnetite cell the $Fe^{\cdot\cdot}$ ions form four layers

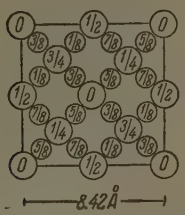

Fig. 1. Distribution of iron ions in an elementary magnetite cell.

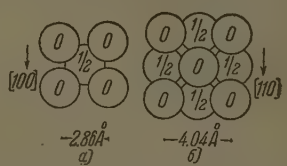

Fig. 2. Structure of α-iron.

having the same structure, located at the heights $^1/_4a$, $^1/_2a$, $^3/_4a$ and $1a$. The distances between the ions in each of such nets along the [100] side are equal to 8.42 Å, and along the [110] and the $\bar{[}110]$ sides, to 5.95 Å.

The $Fe^{\cdot\cdot\cdot}$ ions also form four layers having an identical structure, located at the heights $^1/_8a$, $^3/_8a$, $^5/_8a$ and $^7/_8a$. The distance between the ions in each of such nets along the [100] side is equal to 8.42 Å, and along the [110] and the $(\bar{1}10)$ sides, to 5.95 and 2.975 Å alternately. In metallic iron the two-dimensional (001) nets form a simple square pattern, with the distances

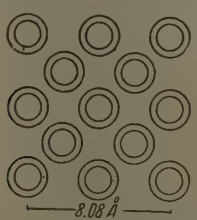

Fig. 3. Displacement of four layers of $Fe^{\cdot\cdot}$ of magnetite by a layer of atomic iron.

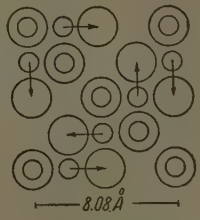

Fig. 4. Displacement of two layers of $Fe^{\cdot\cdot\cdot}$ of magnetite by a layer of atomic iron.

between the iron atoms along the [100] side being equal to 2.86 Å, and along the [110] and the $\bar{[}110]$ sides, to 4.04 Å. An elementary cell consists of two such layers located at the heights $^1/_2a$ and $1\ a$ (Fig. 2, *a*).

A face-centered pattern in the (001) net of iron may be obtained by taking the non-primitive cell which is bounded by the [110] and the $[1\bar{1}0]$ sides (Fig. 2, *b*). Upon comparing this latter pattern with the face-centered pattern of the (001) net of magnetite, it becomes evident that, upon joining the [110] side of magnetite and the [110] side of iron, a complete coincidence of patterns results. With this the deviation between the coinciding parameters is equal to 3.9% ($a=8.42$ Å in magnetite and $2a\pm8.08$ Å in iron), while investigators of the interpenetration of various minerals allow a deviation in the parameters up to 20%.

Let us analyze the mechanism of the displacement of magnetite by α-iron. In Figs. 3 and 4 it is shown how the rearrangement of four $Fe^{\cdot\cdot}$ layers (Fig. 3) and two $Fe^{\cdot\cdot\cdot}$ layers (Fig. 4) of magnetite into a separate layer of atomic iron may take place. As can be seen from the figures, the iron ions

of an elementary cell of magnetite may fill up three layers of atomic iron. With this the co-ordinates of individual atoms should change mainly in the [001] direction.

The elimination of oxygen during the reduction of magnetite leads to the disintegration of the octahedral and tetrahedral polyhedrons of $Fe^{\cdot\cdot} O_4$ and $Fe^{\cdot\cdot\cdot} O_6$. Since the elimination of oxygen from the lattice is accompanied by an increase in the free volume which cannot be compensated by the increase in the radii during the transformation of iron into the atomic form*, the iron lattice should be compressed. The magnetite cell, replaced by six face-centered iron cells (Fig. 5), undergoes severe compression in a direction perpendicular to the [001] axis** which leads to a reduction

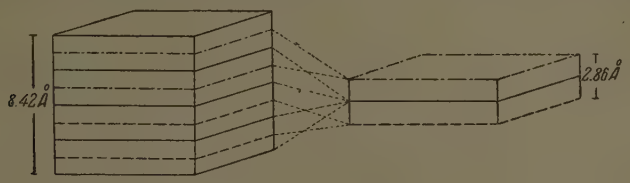

Fig. 5. Diagram of the transformation of the magnetite lattice into a lattice of α-iron.

in the thickness of the displaced layer to one third. During this those insignificant migrations of the iron atoms mainly along the [001] axis which accompany the rearrangement of the lattice, can be easily accomplished.

Thus, in the process of the displacement of magnetite by iron, the volume of the elementary magnetite cells is filled up with three times the number of iron atoms (instead of 24 iron ions, 72 atoms from 18 face-centered cells or 36 simple cells of iron get into the magnetite cell).

Analogously, such a displacement may take place also in the two other cubic plane nets of magnetite. It should be noted that the orientation of the $[0\bar{1}1]$ side representing the densest distribution of ions in magnetite, nearly coincides with the direction of the $[1\bar{1}\bar{1}]$ side of metallic iron, along which the densest distribution of atoms in the structure of iron is preserved.

The explanation of the process of the orientated displacement of magnetite by iron which has been presented above is corroborated by an analogous orientation of the magnetite film which Mehl and McCandless ([8]) obtained during the oxidation of wüstite films.

Conclusions

1. On the basis of a comparison of the structures of magnetite and metallic iron, a theoretical foundation is given to the previously established experimental fact ([1]) of the coincidence of the [001] side of magnetite and the [001] side of α-iron which displaces it.

2. It was noticed that, during such a displacement, the crystalline film of α-iron should orientate itself with its [110] side parallel to the [100] side of magnetite.

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* The radius of $F^{\cdot\cdot\cdot}$ is equal to 0.67 Å and the radius of $Fe^{\cdot\cdot}$ is equal to 0.83 Å, but the atomic radius of iron for an octuple co-ordination is equal to 1.23 Å.

** Such a contraction of the iron lattice, when the displacing layer is very thick, should lead to distortion of the orientation. Experimental data point to this ([1]).

3. The structural side of the mechanism of the displacement of magnetite by iron is analyzed.

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PEDOLOGY

GEDROIZITE IN THE ALKALI-SOILS

By I. D. SEDLETZKY

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It has already been shown (1) that in the colloids of certain soils minerals are encountered which defy identification with any mineral actually known. These minerals have been named „X-minerals".

After having produced an artificial colloid mineral under laboratory conditions and thoroughly studied its composition, properties and structure, the soil colloids may be compared afterwards not only to standards of natural, but also of artificial minerals.

This method seems to be the best way to study successfully the composition of soil pedolites. The importance of synthetic work in the field of soil colloid mineralogy becomes particularly conspicuous if it be considered that organic mineral substances (humus+the alumosilicate part of colloids) are encountered in soils, constituting a class of quite peculiar compounds unknown to classical mineralogy. Experiments on the synthesis of new minerals have been conducted since 1932 under the direction of I. N. Antipov-Karataev. Inasmuch as temperature and pressure are not high in soils, experiments on synthesis are carried on under ordinary room conditions.

From different solution and at different pH of the medium alumosilicates of varying composition are obtained; these are air-dried, transferred into jars with ground-in stoppers and kept for a long time in the laboratory. They are regularly subjected to X-ray structure analysis and heating curves are plotted by the method of differentiation; then they are dehydrated, analyzed chemically and microscopically etc. Usually the new minerals are first amorphous, but afterwards they pass to crystalline forms, which are stable and do not change for a long time.

2. Assuming the existence of special minerals rich in sodium in alkali-soils, the author has obtaided, under alkaline conditions approaching those of the alkali-soil, a synthetic mineral given by us the name of gedroizite (2). Gedroizite contains 13.8% Na_2O and has the following formula: $Na_2O \cdot Al_3O \cdot 3SiO_2 \cdot 2H_2O$, characterized by high sodium content. Besides, the preparation contained 7.3% K_2O, the alkali-content thus amounting to 21%.

Investigation on gedroizite have been carried on for 5 years and are still in progress. The X-ray photograph for gedroizite obtained in 1939 is somewhat different from the original one made for gedroizite in 1937. The differences consist in a more complete crystallization of gedroizite. Towards 1939 a number of lines have appeared which arose in 1937 upon ignition of gedroizite. The principal lines, however, remained unchanged. It may be assumed that in 1939 gedroizite has acquired his ultimate form and its powdergram may be taken as typical of gedroizite.

The most characteristic interference lines for gedroizite are the Debye lines for which d_{hkl}: 3.69; 3.21;1.60 and 1.02 Å will be given respectively.

Among the clay minerals known in literature none would give an X-ray diagram similar to that of gedroizite (Fig. 1); consequently, it is a new mineral.

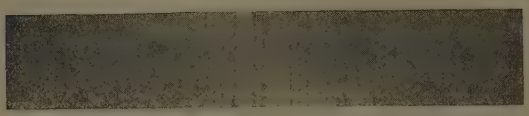

Fig. 1. X-ray photograph.

Among the other constants the heating curve (Fig. 2) obtained by the Le-Chatelier method which is characteristic of gedroizite and the dehydration curve (Fig. 3) should be noted. Gedroizite shows the index of refraction 1.483 ± 0.003 and an exchange capacity of 46.50 mg-equiv. per 100 g.

3. It stands to reason that gedroizite remains an artificial mineral until it is found in soils. As soon, however, as it is detected in the colloids of soils, gedroizite becomes a natural mineral.

It has been mentioned above that the object of gedroizite production was to obtain a mineral characteristic of alkali-soils, i. e. a mineral rich in sodium in an exchangeable and non-exchangeable form. For the last two years we have subjected to investigation colloids of alkali-soils of different stages from the left bank of the Volga, Middle Asia, West Siberia,

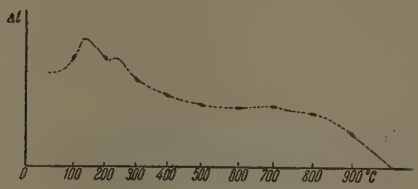

Fig. 2.

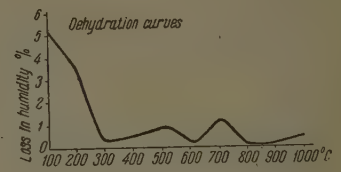

Fig. 3.

the Chernigov region and from other places of the Union. Two fractions of soil colloids have been studied: <0.2 μ and 0.2—2 μ. In some cases the fractions: <1 μ and <2 μ have been studied. The alkali-soils studied comprised soda, chloride-sulphate, alkali-soils and those transformed into steppes ([3]). In most cases the alkali-soil colloids (0.2 μ) consisted of montmorillonite, beidelite and muscovite. However, X-ray diagrams showed frequently, especially in soda alkali-soils, several lines (more frequently one line with $d = 3.21$—3.25) which could not be put down to the presence in the colloids of any mineral known.

As regards the fractions from 0.2 to 2 μ, they had in some cases a mineralogical composition similar to that of smaller fractions, and in other cases (namely, in soda alkali-soils) the composition of minerals was sharply different. The difference consisted in the presence of a number of other minerals: kerolite, sericite etc. Colloids from the alkali-soil horizon of the soda alkali-soil contained gedroizite. The presence of no other mineral in the colloids of the alkali-soil horizon is clearly seen from the interpretation of the X-ray diagram (Table 1).

The interference lines of the X-ray photograph put down to gedroizite cannot be referred to any of the clay minerals known, while all the lines in the radiograph of the alkali-soil colloids fit fairly well within the X-ray spectrum characteristic of gedroizite. If we had no X-ray diagram of gedroizite at our disposal, all these lines, as being unidentified, should be put down to the presence in the colloids of some unknown mineral. The question suggests itself, if in this case we had possibly to do with an accidental coincidence of lines.

To prove the appurtenance of this line to gedroizite it is impossible to have recourse to thermal analysis, because the heating curves of gedroizite and montmorillonite approach each other rather closely. The determination of the indices of refraction is no safe criterion either. The fact of preservation of the gedroizite structure when heated above 1000° C may be taken as a final proof. Experiments made with ignited colloids of an alkali-soil from the Chernigov region showed a complete preservation of the X-ray diagram of gedroizite.

Now the question arises whether gedroizite is a mineral of alkali-soil. If gedroizite be a soil mineral, then it should be absent in the parent rock. A study of X-ray photographs from other genetic horizons and of the parent rock from a section of the same alkali-soil showed gedroizite to be present only in the alkali-soil horizon. Above and below gedroizite is absent. Thus, the appearance of gedroizite in the alkali-soil horizon is closely connected with the origin of these soils and particularly of the alkali-soil horizon. As it is known the alkali-soils originate from salt-marshes in result of a desalinization of the latter. Readily salts are washed out of the upper horizons.

Table 1

Interpretation of the Powdergram of a Soda Alkali-Soil (0.2—2μ)

Fraction 0.2—2 μ		Gedroizite (artificial)	
d, Å	Intensity	*d*, Å	Intensity
3.54	medium	3.60	medium
3.47	» *	—	—
3.26	very strong	3.21	very strong
3.11	weak	—	—
2.94	strong	2.96	weak
2.77	weak	2.70	»
2.52	medium	2.56	medium
2.42	»	2.39	»
2·23	»	2.24	»
2.04	»	2.03	»
1.955	»	1.98	very weak
1.84	»	1.83	weak
1.786	»	1.78	»
1.639	weak	1.69	very weak
1.572	very weak	1.57	medium
1.518	medium	1.51	»
1.484	»	1.47	»
1.393	weak	1.36	weak-medium
1.356	strong	1.34	medium
1.325	very weak	1.31	medium-weak
1.273	weak	1.27	weak
1.182	»	1.23	»
1.165	»	1.20	wery weak
1.140	»	1.16	»
1.110	very weak	1.12	»
1.088	»	1.08	»
1.037	»	1.05	»
1.026	»	1,02	medium
0.994	»	1.00	very weak
0.968	»	0.957	»

The absorbing soil complex is saturated with sodium. Soda formed at the expense of exchangeable sodium provides a highly alkaline medium. In consequence a highly condensed (during the dry season) columnar horizon named the alkali-soil horizon appears in the upper horizons of the soil.

In the lower horizon it is absent, owing to the presence of readily or sparingly soluble salts. The uppermost crust of soil is also devoid of the properties of an alkali-soil horizon, owing to a better washing out and, consequently, a lower exchangeable sodium content. A maximum amount of exchangeable sodium is contained in the columnar horizon and the reaction of the medium is highly alkaline (Table 2).

Here in an alkaline medium and in the presence of large amounts of sodium all the conditions are favourable for the formation of gedroizite. And indeed gedroizite originates from the alkali-soil horizon. Other horizons of the soil are lacking such favourable conditions: the alkalinity of the medium is lower, the amount of exchangeable sodium less, etc. And gedroizite is absent there. Consequently, gedroizite is a mineral of alkali-soil horizons, and it concentrates in the fraction 0.2—2 μ (Table 3).

* Part of the lines not identified and belonging to another mineral.

Table 2

Exchangeable Bases and pH of the Soda Alkali-Soil of the Chernigov Region

Horizon	Depth of sampling, cm	Total alkalinity	pH	Exchangeable bases in mg-eq. for 100 g	
				Na	Ca+Mg
Eluvial	0 — 0.5	0.366	10.30	1.44	20.43
Alkali-soil	0.5— 10	0.478	10.40	7.53	26.18
	24 — 30	0.235	2.25	4.67	33.29
	60 — 70	0.094	9.14	5.40	35.45
	190 —200	0.064	8.80	0.75	25.35

4. It is still not quite clear why gedroizite concentrates in the larger fraction (0.2—2 μ) and is absent in the smaller one (<0.2 μ). It is true that in the soda alkali-soils of the left bank of the Volga the X-ray photographs of the fraction 0.2 μ show the lines d=3.21 Å. This line is most intense in the X-ray photograph of gedroizite. Inasmuch as in the X-ray photographs of alkali-soil-colloids (0—7 cm) this line is fixed alone and is moreover of slight intensity, it should be assumed that gedroizite is present in very small quantities (3—5%). The remaining 95% consist of montmorillonite.

Table 3

Composition of Pedolites of the Soda Alkali-Soil of the Chernigov Region

Horizon, cm	Fraction 0.2—2 μ	Fraction <0.2 μ
0 — 0.5	montmorillonite, sericite	montmorillonite
0.5— 10	gedroizite	montmorillonite, muscovite
24 — 30	kerolite	Mg-beidelite
60 — 70	»	kerolite, montmorillonite
190 —200	kerolite, montmorillonite	montmorillonite

The above data permit of the following conclusions:

1) About the finding of gedroizite in the soda alkali-soil colloids as a specific mineral constituent of the absorbing soil complex. Gedroizite is a specific mineral of soda alkali-soils. The presence of a series of other specific minerals in other alkali-soils may be assumed. The alkali-soil series of soils is characterized by alkali-soil pedolites distinguishing the alkali-soil from all other types of soil. They might be named solonzerites.

2) The composition of the pedolites of alkali-soils is intimately connected with the origin of soils. The mineralogical composition of individual genetic horizons is strictly correlated with the type of the soil-formation process.

3) In the course of the soil-formation process peculiar and new minerals are formed which are in a highly disperse (colloidal) state. Gedroizite is but one representative of this class of minerals.

4) The issue of our methodical conclusion will be that the synthesis of minerals is an essential and indispensable part of the study of the mineralogical composition of soil colloids.

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GENETICS

DIFFERENCE IN BLOOD VALUE OF KARAKUL SHEEP IN CORRELATION WITH THEIR ORGANIC CONSTITUTION

By E. P. PANFILOVA

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The major part of the data to be found in literature on the blood value of farm animals are mainly confined to the establishment of definite intervarietal differences ([1–4]). Recently papers have appeared dealing with blood composition of animals within the same variety, whereby attempts were made to establish connexion between blood value and productivity ([5–9]).

Gärtner and Heidenreich ([5]), who have studied the dry substance of the blood in Merinos, found a correlation between this blood factor and the body weight as well as with the yield of wool. According to Kushner ([7, 8, 9]) there is a connexion between the blood components (haemoglobin, erythrocytes), on the one hand, and body weight and yield of wool, on the other; there are also certain differences between the Würtemberg and the Gunib breeds. Geske ([6]), while studying the blood composition in Karakul sheep, found a correlation between the size of red blood corpuscles and the animal's body weight.

In our present work we set ourselves the aim of studying the haemoglobin value and size of the erythrocytes in Karakul sheep. Our investigation was made at the «Nachalo» collective farm in the Ak-Mechet district of the Crimean Autonomous Soviet Socialist Republic. Our material comprised 59 dams, two years of age. The examination of the blood was made in the spring at the end of April, 30 days after yeaning. Blood was taken from the ear vein in the morning before the sheep were taken to graze. At the examination the quality of the wool and the constitution of each dam were recorded. Owing to technical difficulties, the body weight was ascertained only for 38 dams. The haemoglobin value was determined by the Sahli method and the diameter of the erythrocytes by dry undyed smears.

Haemoglobin Content. The average haemoglobin value in Karakul dams was found to equal 52.23 ± 0.83 (from 35 to 68) in Sahli units. According to the data given by Geske ([6]), the haemoglobin value in Bokhara Karakul sheep equalled 61.17, varying between 40 and 74. The higher haemoglobin value given by Geske may be explained by the fact that this was a heterogeneous group of sheep both as regards sex and age, 12 of the 66 animals investigated being rams. In regard to sexual dimorphism the data given by Kushner point to a considerably higher blood value in rams. In respect to age the sheep examined by Geske varied between one and 10 years. Moreover, the examination was not done in one season but dragged

out from March to September. The average body weight of the Karakul sheep examined by Geske equalled 46.5 kg as compared with 38.88±0.64 of our animals. The haemoglobin value of particular animals varies considerably. The index of variation in haemoglobin is high and equals 12.5%.

Dams of the minus variant for haemoglobin content proved to be of lighter body weigth, a difference of 3.5 kg in body weight between two groups of dams is significant. In support of this we have calculated the coefficient of correlation between body weight and haemoglobin value and found it to be 0.375±0.13.

Thus, contrary to Kleeberg (²) who failed to find any connexion between body weight and blood value, our material enables us to say that the coefficient of correlation is sufficiently high and the figures highly significant ($r=+0.375+0.13$). This testifies to the regularity of the correlation between body weight and haemoglobin value.

For the purpose of studying the connexion between haemoglobin content and the constitution of the animals we have divided our material into three groups. One succeeds in dividing the Karakul dams into three rather distinct groups according to their exterior, body weight, thinness of skin and quality of wool.

The first group is of a coarse type: the dams, with a coarse and heavy skeletal growth, have an average height of 62 cm at the withers, length of trunk is 67 cm, depth of chest 29.5 cm, body weight averages 45 kg. The growth of wool is insignificant. The fleece of the coarse animals contains only a little fluff; it is coarse (50 microns), not of a wavy structure and grows rapidly after shearing. Length of wool averages 10 cm.

The second is of a slender type; the dams are of a slender skeletal growth; their height at the withers averages 60 cm, length of trunk 65 cm, depth of chest 28 cm, body weight averages 39 kg. The growth of wool is considerably higher; the wool is fine (28 microns). The fleece of this type of sheep contains a great deal of fluff, the wool is short and falls in the wavy plaiss. The wool averages 5 cm in length.

The third, normal type of animals, holds an intermediate position. These are proportionally built animals with a solid skeletal growth. The fleece of these sheep contains a medium quantity of fluff. The wool averages 40 microns in fineness and is 8 cm long. The difference in the haemoglobin value of these three groups is shown in Table 1.

Table 1

Haemoglobin Content in Sahli Units in Correlation with Constitutional Type of Dams

Group	Haemoglobin				
	n	*M*	±*m*	σ	*C*
Slender	8	44.75	1.96	5.55	12.83
Normal	27	48.89	0.80	4.17	8.52
Coarse	24	57.13	0.85	4.20	7.35

From Table 1 it is evident that the average haemoglobin value of a dam of the slender type is less than that of both normal and coarse type dams; the haemoglobin value of the coarse type dam exceeds that of the slender type dam by 12.38±2.16 and that of the normal type dam by 8.24±1.16; in both cases the difference is significant. The haemoglobin value of dams of the normal type exceeds that of the slender type dam by 4.16±2.11, but this difference is less than its triple error. Under the conditions prevailing at the Nachalo collective farm in Crimea the coarse type of dams proved more

adapted to life and less susceptible to disease. Thus, the percentage of animals affected by scabies and helminths is higher among dams of the slender type than among those of the coarse type; cases of death among dams of the coarse type totalled 1.5% in 1938 as compared with 2.5% among the dams of the slender type. The effect of the higher haemoglobin value is better health and greater resistance on the part of the animals to adverse external conditions. However, inasmuch as the dams of the coarse type had on the average a greater body weight, too, it is possible that the differences in haemoglobin value between animals of different types are determined rather by this difference in body weight.

Size of Erythrocytes. Fifty erythrocytes of each smear were measured. On the basis of the averages for the particular animals an average was deduced for the whole breed. This average has found to equal 4.54 ± 0.053 microns and fluctuated between 4.13 and 4.84 microns. The coefficient of variation in the size of the erythrocytes is inconsiderable, it equals 2.78%; thus, different animals differ but little as to the size of their erythrocytes. For the sake of comparison we give in Table 2 the size of erythrocytes in dams of different breeds according to data produced by other authors ([7],[6],[1]).

Table 2

Breed	Size of erythrocytes		
	M	$\pm m$	n
Kushner's data			
Local Gunib dams	5.16	±0.06	9
Pure blood Würtemberg dams	5.28	±0.07	31
Cross F_1 » »	5.20	±0.06	31
» F_{2b} (3/4 blood) »	5.22	±0.06	35
Geske's data			
Bokhara Karakul sheep	4.946	—	56

Moreover, Götze points to the fact that Karakul dams proved to possess erythrocytes of the smallest size, while erythrocytes of the largest size were found by him in Rambouillet, Merinos, Hampshires and Ostfrieslands. Comparing the size of erythrocytes in different sheep varieties, we find that on the average Karakul dams have erythrocytes of the smallest size, while all the other varieties mentioned have erythrocytes of a larger size. We do not cite here Bogolyubova's data on the size of erythrocytes in Hampshire, Wallachian and other varieties, since this author applied a different method of fixation (vapours of osmic acid). Table 3 shows differences in size of erythrocytes in correlaton with the body weight of sheep.

Table 3

Size of erythrocytes	Body weight				
	M	$\pm m$	σ	C	n
Up to 4.4 micr.	39.5	1 44	4.56	11.54	10
From. 4.4 to 4.6 micr.	39.62	0.89	3.46	8.74	15
» 4.6 to 4.84 »	42.58	1.09	3.64	8.54	11

Dams with larger erythrocytes have also greater body weight than dams with small erythrocytes. The body weight difference between the two extreme groups of dams arrayed according to size of erythrocytes equals 3 kg. The coefficient of correlation between body weight and size of erythrocytes equals

$r = +0.284 \pm 0.153$. This goes to show that there is a small positive correlation between this blood factor and the body weight of the animal; it is, however, not significant.

We have in addition investigated the differences in the size of erythrocytes of different types of animals (Table 4).

Table 4

Size of Erythrocytes in Karakul Dams in Correlation with Their Constitution

Type of dam	Size of erythrocytes				
	n	M	$\pm m$	σ	C
Slender	8	4.38	0.064	0.159	3.0
Normal	27	4.49	0.042	0.120	2.21
Coarse	22	4.59	0.053	0.138	2.50

The erythrocytes of the coarse type dams are by 0.10 ± 0.064 microns larger than those of the normal type dams and by 0.21 ± 0.078 microns than those of the slender type dams. Although on account of paucity of the material in the slender group the difference is not quite significant, the absolute differences are nevertheless quite considerable. This offers us ground to maintain that animals of a coarser type have larger erythrocytes. In this connexion it is of interest that Kleeberg (2) also points out that lowland sheep with a slender constitution and live spirits possess small erythrocytes of a higher haemoglobin value, while the meat breeds with their coarse constitution possess larger erythrocytes.

Thus, our data on haemoglobin content and size of erythrocytes in Karakul sheep show that: 1) dams with a greater body weight have a higher haemoglobin value and, vice versa, dams of the same age with a lighter body weight have a lower haemoglobin value. The coefficient of correlation between these two factors in a group of 38 dams at the age of two years equals: $r = +0.374 \pm 0.13$, while the coefficient of correlation between the size of erythrocytes and body weight equals $r = +0.284 \pm 0.153$; 2) a certain connexion may be traced between the blood picture and the animal's constitution: the coarser the dams, the greater the size of their erythrocytes and the higher their haemoglobin value; 3) the study of the blood value may serve as an additional criterion for determining the constitution and evaluating the breed qualities of the animal.

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GENETICS

VARIATION OF THE SHAPE OF THE «KURDJUK» AND OF THE TAIL APPENDIX IN TUSHIN SHEEP

By M. D. RCHEULISHVILI

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As the characters of the tail are of taxonomic importance, it is of interest to analyze in what measure does the classification of Pallas-Nathusius-Chirvinsky determine the real taxonomic position of the Tushin sheep and, moreover, to what extent the elements of the various characters of the Caucasian breeds of sheep are represented in the structure and shape of the «kurdjuk» of the Tushin sheep.

Any form of the «kurdjuk» in the Tushin sheep possesses two symmetrical fat-cushions separated from each other from the inferior (hairless) side by a rather deep furrow. On the upper surface of the «kurdjuk» grows ordinary fleece wool which differs from the main assortments only in coarser hairs.

By the shape and length of the tail the Tushin sheep fall into 2 basic types: «dumeuli» and «kuntuli». Between the extreme well separated variants all possible intergradations are observed. The tail in «dumeuli» (Fig. 1, right) is more hanging, friable and long; in extreme cases it reaches the hock. Disregarding the additional cushion (which is observed in reduced size in the Tushin sheep, too), some forms of this type approach in shape and hanging of the tail to Mazekh-balbas. In the type «dumeuli» the tail is hanging irrespective of the degree of fulness. The other type of the «kurdjuk» in the «kuntuli» is distinguished by elevated position and is almost at the level of the sacrum. It is more compact as compared to «dumeuli». The horizontal position of the «kurdjuk» of this type does not depend on the amount of fat deposited. Irrespective of the tail appendix this type of tail is similar to the «kurdjuk» of the Kurdjuk sheep (Fig. 2, left).

In the Tushin sheep two sharply distinct types of «kurdjuks» are thus met with; one extreme is similar to the tail of the Mazekh, the other—to the genuine «kurdjuk» of the Kurdjuk sheep.

Out of the total of 726 females of the Tushin sheep 62.26% had tails of the «dumeuli» type, whereas the remaining 37.74% had «kurdjuks» of the «kuntuli» type. The «dumeuli» type thus proved to be more common in the Tushin sheep than the «kuntuli» type; this may be explained both through selection for «dumeuli» (?) which was practised by certain sheep-breeders and through dominance of the type «dumeuli» over «kuntuli». According to our data (unpublished), the shape and length of the fatty tail of Mazekh sheep is completely dominant at the lamb age; in adults it is partly dominant (owing to age variation) over the «kuntuli» type of the Tushin sheep. Clear dominance of the «dumeuli» type over the «kuntuli» type is also observed in crosses between pure-bred Tushin sheep.

The «kurdjuks» of the I and II type may be either closed, i. e. showing no hairless underparts, or open. The open hairless underparts («duma-akudebuli» type), owing to the elevated position of the tail appendix have a triangular shape (Fig.1, right, and Fig.2). The hairless underparts of the other type «duma-kaltiani» are inconspicuous from the outside (Fig. 1, left, and Fig. 3).

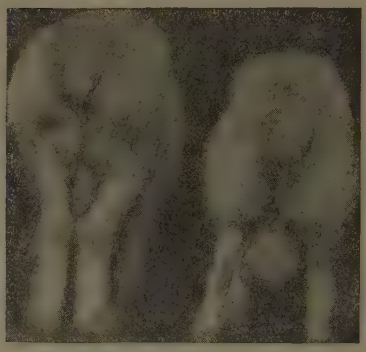

Fig. 1. Adult Tushin rams.

All the enumerated variants have free (in the majority of cases non-fatty) tail appendices. The «kurdjuk» in the Tushin sheep is strongly depressed from the upper side at the last third of its length; further the tail is strongly elevated, in some cases («kuntuli» type) reaching the level of the sacrum, then the above-mentioned part of the tail is bent downwards and becomes thinner, forming the non-fatty part of the tail appendix. Out of tens of thousands of sheep examined by us only four had no tail appendices.

The tail appendix of normal curvature from the depressed region of the «kurdjuk» rises up and, after becoming free, bends downwards and hangs straight parallel to the longitudinal furrow of the «kurdjuk» (Figs. 1 and 3). Out of the 726 females 74.12% had tail appendices with such curvature. In some individuals the tail appendix is strongly bent to the left or to the right at the very transition point to the free end (Fig. 2), the majority of animals, however, showing a slight curvature.

Fig. 2. One and a half year old Tushin rams.

Fig. 3. One and a half year old Tushin rams.

In order to find out whether the curvature of the tail appendix is a hereditary character, we have arranged crossing which demonstrated (not yet published) the dominant nature of this character. A Tushin ram № 225.E. (Fig. 2) possessing strong curvature of the tail appendix to the right was mated during 3 years with females having normal or bent to the right tail appendices. In all cases the progeny had either strong or slightly curved tail appendix. It should be noted that some of the rams clearly showed segregation, whereas some of them have not at all transmitted the character

in question. The above-mentioned phenomenon enables one to the conclusion that aside from sheep having hereditary curved tail appendix there are such, in which it is of non-hereditary nature (embryonic mutilation, etc.).

Aside from the types enumerated above, such are met with among Tushin sheep which possess tail appendices of normal curvature, but thick and fat (fatty) instead of being thin, like in preceding types. This type is characterized by gradual growing thinner from the base to the tip, which results in a triangular shape (Fig. 3). This type of tail appendix has the appearance of an additional fatty tail formed above the «kurdjuk» which makes it similar to the corresponding structure in Mazekh-balbas sheep.

Summing up, we arrive at a conclusion that the majority of the Tushin sheep possess «kurdjuks» of the «dumeuli» type, which is similar in its extreme form to the fatty tail of the Mazekh-balbas. One should not conclude from this, however, that Mazekh or Balbas had strong influence upon the formation of the modern type of Tushin sheep. As shown in our preceding paper ([7]) Mazekh and Balbas could be crossed more or less extensively with the Tushin sheep only in the second decennial of the XX century; it should be added that in the majority of cases, especially at large farms, pure Mazekhs were not crossed with Tushin sheep (or only occasional crosses were made for production of coloured wool) owing to inferior wool and meat qualities.

Among all the breeds existing in Transcaucasia the Tushin sheep are most similar to Lezgin, Bozakh and Karabakh sheep in the shape of the «kurdjuk»; the influence of these breeds upon the Tushin sheep, as well as the mutual influence of the latter upon them is proved by a number of other characters and by historical data ([7]).

In Tushin sheep fat is deposited on the sacrum and thighs, as well as around the bones of the tail. The «kurdjuk» is well developed in width. They show, therefore, a certain similarity to the genuine kurdjuk-sheep (disregarding the number of the tail vertebrae which are more numerous in the Tushin sheep than in the kurdjuk sheep); it differs in some cases only in the tail appendix which is observed in the kurdjuk-sheep only as a rare exception ([1–6]).

On the other hand, as to the tail skeleton, presence of a free tail tip and deposition of fat in the tail itself, the Tushin sheep show a great similarity to fatty-tailed sheep. The Tushin sheep and the like (Karabakh, Bozakh etc.) are a sort of connecting link between the Kurdjuk sheep and fatty-tailed sheep.

This statement prevents their including into the kurdjuk-group or into the group of the genuine fatty-tailed sheep as done in the classification of Pallace-Nathusius-Chirvinsky, even with addenda by Ivanov ([3]).

The nature of things demands their isolation as a distinct group, as done by Prof. Bogoljubsky ([2]) who placed them (together with breeds similar in the number of vertebrae) into a distinct group hemisteatopyga.

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GENETICS

VARIATION IN TAIL FORM IN IMERITIAN SHEEP

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The length and form of the tail in sheep, along with the development of fat, are underlying the existing classification of Pallas-Nathusius. Hence a study of tail variation in the existing breeds, especially among the old races, is of some interest.

The Imeritian sheep are distributed at present over the whole of Western Georgia, not including the regions of the Autonomous S. S. Republics of Abkhazia, Adzharia and some districts of Mingrelia. In Abkhazia the Karachaevo sheep is widespread, while in the latter two the so-called Adzharian breed is prevailing which is believed by some to present a Turkish breed, coming from regions adjoining Adzharia (in most of the Mingrelian districts prevails the Imeritian sheep).

The data of our investigation (7) warrant the contention that the Imeritian sheep is not pure-bred throughout the entire range of its distribution, being rather mongrel as a result of crosses of different strains, and interbreeding with the Tushin and Adzharian sheep, and in some regions—with the Karachaevo and Ossetian breeds as well. The Chiatury district being more isolated, not only the native sheep, but also the cattle and swine encountered there are relatively true-breeding. The sheep which are now widespread in the Chiatury district, extended their range over the whole of Eastern Georgia till about the XI century, in places persisting there up to the second decennial of the XX century(7). There were found Georgian bas-reliefs, tomb-stones, and all kinds of miniatures exhibiting figures of sheep resembling in the form of horns and tail, and a number of other characteristics, the Imeritian and old-Assyrian sheep(7). All this material evidence and descriptions taken from old records(2), the history of the Georgian people(4), as well as the study of the sheep population of Western Georgia, justify our conclusion in regard to the common origin of the Imeritian and old-Assyrian sheep, and greater or lesser degree of pure-breeding of the sheep from the Chiatury district.

According to the classification of Pallas-Nathusius-Chirvinsky supplemented by Ivanov(5), the Imeritian sheep along with the Valachian (*Ovis strepsiceros*) and Malich breeds is placed in a group of sheep possessing a long and fat tail with a straight non-curved tip. How true this classification will prove to be in respect to the Imeritian sheep, will be seen from further evidence; at this stage let us note their great variability even in the regions of their true-breeding.

By the form of the tail the sheep of the Chiatury district fall into three principal groups: the first includes sheep with tails of the type № 147 (Fig.1). In these sheep there is no transition from the fat to the lean portion of the tail, the tail showing a marked swelling at the very base and terminating in a pointed wedge. Another variation of the above type differs from this one but in the bluntly pointed triangle, i. e. the fat deposit continues to the tip of the tail (Fig. 1, № 113). The third variety of the same group is quite similar to the main type, differing from it in a much smaller (by nearly 30%) tail length (Fig. 1, № 64).

To this group is likewise referred by us the form of a tail whose fat portion is very much like the main type, the only difference between them being the presence in the former of a curve at the base of the lean portion of the tail (Fig. 1, № 155).

This group of tail types bears likeness to that of the tail varieties of the Valachian sheep.

The first group constituted 45.5% of the entire population of the investigated sheep of this region. This group comprised 38.1% of the main type, 36.9% of the second subtype, 15.2% of the third and 9.8% of the fourth.

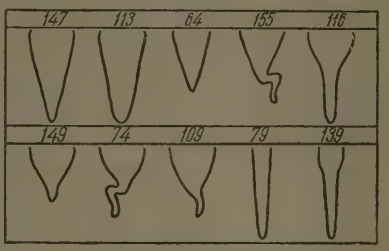

Fig. 1. Variation of tail form in Imeritian sheep.

According to the range of distribution, the next place (34.6%) belongs to the type of tail in form very much alike that which is quite common in the Valachian sheep ([3]). From the preceding type it is distinguished by the oval form of the fat portion of the tail with lateral flattening above its median portion, marking the beginning of the lean tip of the tail (Fig. 1, № 116). The lean tip of this type of tail (53%) takes up on the average 65% of the total length of the tail. The next variety (25.7%) of this type is quite similar in form to the main type, differing from it but in the greatly shortened lean portion of the tail. The length of the latter is about 27.3% of the total length of the tail (Fig. 1, № 149). Further varieties (20.1%) of this group (Fig. 1, № 74 and 109) likewise resemble in the form of the fat portion of the tail the main type differing from it but in the curve of the lean part of the tail.

The third group of sheep (19.9%) possess a tail essentially similar in form to the lean tail of the Merino breeds (Fig. 1, № 79), some of its varieties having a small fatty deposit at the base of the tail (Fig. 1, № 139) which could have developed through crossbreeding of this group with sheep of the preceding groups. However, in order to explain such a slight swelling, perhaps, we need not necessarily admit the possibility of hybridization with the preceding types, considering that according to Prof. Adametz([1]) an identical fattening of the base of the tail is observable in Merino sheep.

In the remaining districts of Western Georgia([7]) with obviously hybridized sheep flocks still greater variation in the form of the tail is observable. The tail type most common here (55%) is № 116 (Fig. 1).

In regard to frequency of occurrence, next follows the wedgeshaped tail form of the type № 147 (Fig. 1), along with the tail form of Tushin sheep known under the name of «kuntuli» (18.6%), though much inferior in size to the (fat rump) «kurdjuk» of the Tushino strain. Besides the «kuntuli» type, there may be encountered another type of «kurdjuk» of the Tushin strain, strongly reduced but similar in form (though somewhat narrow and elongated) known as «dumeuli» (4.1%). In the same districts occur sheep

with tails exhibiting a bare underpart along the entire length, due to the raised condition of the appendix and uncovered fat portion well cushioned with fat. We feel free to assume that this variety too has arisen under the influence of hybridization of the Tushin sheep, since it is a character rather common in the latter and is called «duma-akudebuli».

Beside the type of tails of Tushin sheep, to a smaller extent (3.2%) are met sheep with the type of tail peculiar to the Karachaevo and Karakul breeds. Thus, in the districts of unsystematic hybridization there are more varieties of tail form in Imeritian sheep than in the Chiatury district. According to the tail form in districts of promiscuous hybridization is clearly felt the influence of the Tushin breed, in the same manner as it has been proved for a number of other characters(7). Many varieties of the tail form could have arisen by way of inbreeding inter se and interbreeding between the Tushin-Imeritian, Imeritian-Adzharian and complex hybrids.

It is worth mentioning that in tail form the Imeritian sheep generally resemble the Valachian breed, whereas in tail length these breeds differ sharply from one another. While the tail of the Imeritian sheep does not reach to the hock (except in isolated cases), the main bulk of Valachian sheep possess a tail reaching below the hocks, often extending down to the bottom of the foot. According to Prof. Vasin(3), the length of the tail in the Valachian sheep is on the average 43.02 cm, while, according to our own measurement, in the Imeritian sheep it equals 17.67 cm. In Valachian sheep the tail length varies from 27 to 58 cm, and in the Imeritian breed from 6.8 to 25.9 cm, so that the maximum range of the Imeritian does not come up to the minimum range of the Valachian breed. The ratio of the tail length to the height at the withers in Valachian sheep is on the average 69.1% and in Imeritian 33.2%. Hence, according to the form of the tail, the majority of the now widespread Imeritian may be united in one group with the Valachian breed, however, if one should be guided by the length of the tail, the Valachian sheep raised in the USSR are obviously long-tailed, while the Imeritian breed should be placed among short-tailed, or more properly, sheep with a medium-long fat-tail.

In most cases the fat portion of the tail in Valachian sheep (judging by the photos of Vasin) is longer, and occasionally equal to or shorter than the lean portion. In the Imeritian breed the fat portion is on the average about 7.23 cm, and the lean part 11.44 cm long, although there is a great variety in this respect as shown in the table.

Groups according to tail length	Chiatury district	Zestafon district
Fat portion of the tail constituting up to 50% and less of the lean part	41.6%	15.55%
Fat portion of the tail smaller that the lean one, however exceeding 50% of the latter	37.6%	26.80%
Fat portion equals lean portion of the tail	15.2%	46.65%
Fat portion longer than lean portion of the tail	5.6%	11.06%

As seen from the above table, in the district of true-breeding of the Imeritian sheep (the Chiatury district) the fat portion is in most cases (79.2%) shorter than the lean part of the tail, whereas in districts of hybridization the aspect is different.

By comparing the tail variations in hybrids of the first and second generations, upon crossbreeding between the Tushin and Merino strains*, we

* Data not yet published.

arrive at the conclusion that they exhibit all the variations peculiar to Imeritian sheep. On the other hand, the occurrence in the Chiatury district of lean-tailed sheep, especially since we have not accomplished the investigation of hybridization of sheep of this district with lean-tailed strains(7), leads to the conclusion that originally the Imeritian sheep were lean-tailed even in places of their present habitat in Georgia; whereas the fat-tailed varieties sprang up as a result of hybridization of these sheep with the Tushin and partly with the Adzharian breeds. This contention is supported by the known geographer Vakhushti(2), mentioning in his work on the geography of Georgia the lean-tailedness of Imeritian sheep. The above is likewise borne out by studies of ancient relicts of Georgian culture, among which are often met lean-tailed but typical Imeritian sheep identified by the form of their horns.

When comparing these sheep with those on the bas-reliefs of the Numrud palace, representing old-Assyrian rams(6), at first glance a certain discrepancy is revealed. Rams shown on this bas-relief are in their head shape, as well as in form and length of ears, curvature and form of horns, also in extension of the body and structure of extremities, quite similar to the Imeritian ram, though differing from the latter by their manifestly fat tail, whereas the Imeritian strain, as indicated above, possessed a lean tail. We are interpreting this phenomenon(7) thus: the above-mentioned bas-relief is dated 745 B. C., while the Georgian tribes emigrated from their former home (adjoining localities inhabited by Assyrians) in Asia Minor to Transcaucasia a long time before 745 B. C.

The above is conclusive evidence that the fat tail in Assyrian sheep has evolved after the immigration of the Georgian tribes to Transcaucasia, otherwise this character, which is of great economic value, would have been fixed among the Imeritian sheep as well.

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PLANT PHYSIOLOGY

YAROVIZATION OF SEED OF AGRICULTURAL PLANTS BY IMMERSION IN WATER

By I. A. FILIPPENKO

(Communicated by A. N. Bach, Member of the Academy, 31.III.1939)

The works of Lyssenko have shown the development of plants to consist of different stages. In order to make the plant pass any of those stages, a definite combination of various factors is necessary. The stage of yarovization will not take place unless certain conditions of temperature, humidity and aeration are available. If this complex of factors is disordered, yarovization proceeds very slowly if at all.

The experimental data discussed in the present paper have been obtained in attacking the problem as to how the plant will pass the yarovization stage, when the complex of indispensable factors is disturbed by immersing the seed in water.

Seeds of winter wheat Ukrainka in amounts of 5—10 g were put into flasks of 150 cm^3 capacity. After 100—120 cm^3 of tap water had been added, the flasks were stopped with corks so that a small volume of air remained above water. They were placed in a refrigerator ranging in temperature between 2 and 5° C. 24 hours later the water was poured out and the same volume of new tap water poured in, the flasks being put again in the refrigerator. Every 10 days a new portion of seed joined in the experiment and toward the day of planting (April 29) there were 8 kinds of seed varying in time they had passed in water between 10 and 80 days.

3—4 days from the outset of yarovization the embryos showed obvious growth and after 7—8 days in water they ruptured the seed coat and well developed seminal radicles pushed out. In that condition the embryos remained until planting, making no further growth. At the same time the seeds of Ukrainka were yarovized according to Lyssenko (usual yarovization). By the day of planting there were 5 variants ranging in yarovization time between 10 and 50 days.

May 3 all the variants were planted into earthenware pots with threefold iteration. During vegetation period the plants were kept under greenhouse net. Besides Ukrainka a spring variety, *Erythrospermum* 82/02, was involved in the test. The seed of the latter was yarovized in water at 3°C for 80 days. Controls of that variety were yarovized for 20 days according to the usual procedure or imbibed in water a day before planting.

Phenological observations have shown that all plants of spring variety *Erythrospermum* passed the various stages of development simultaneously, irrespective of their previous treatments. Among winter variety Ukrainka

the plants from the seed yarovized according to the usual method for 50 days produced heads in the first half of July. Plants from the seed yarovized in water were tillering throughout June, July, August and the first half of September (Fig. 1).

Toward the end of vegetation experiments, i. e. September 10, the plants treated 40 days in water produced heads, while in plants with other yarovization lengths the growing cone began differentiating. After harvesting the plants, September 25, the cones were measured.

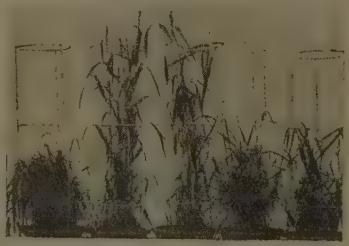

Fig. 1. *1*—from dry seed; *2*, *3*—from 50 days treated according to Lyssenko; *4*, *5*—from yarovized in water for 80 days.

On the basis of data obtained one may draw a biological curve of embryonic spike differentiation in terms of cone length. The maximum in this curve is with the seed yarovized in water for 40 days.

The seed treated in the usual way during a period of 20 days produced plants which came into ear September 20. We thus see that the plants have passed the same length of vegetation period regardless whether their seed was immersed in water for 40 days or given ordinary yarovization treatment during 20 days (Fig. 2).

The conclusion to be drawn from the above facts is that keeping in water (yarovization of winter wheat seed Ukrainka at 3°C) has led to delayed development compared to that of plants yarovized after the usual method. This effect has, however, failed to appear in spring wheat plants *Erythrospermum* which showed equal development whether their seed had been kept in water or yarovized in the usual way or subjected to no such treatment at all.

Fig. 2. *1*—unyarovized seed; *2*—seed treated for 20 days in the usual way; *3*—seed yarovized in water during 40 days.

Taking into consideration the results of phenological observations on wheat's development early in July, we have deemed it wise to complete our experiment by testing other crops for their response to immersion of seed in water. Among the plants involved in the experiment most were known to respond to yarovization positively, say lupine, winter vetch, but with regard to some, viz. winter barley Viner and winter oats, no data whatever on their response to yarovization were available. The seed was yarovized either in water at 3° C for 20 days or in the usual way under the same conditions of time and temperature. Planting was done July 20 into earthenware pots. Exception was made for winter vetch which had its treatment, whether in water or in the usual way, extended over 40 days and was planted much earlier, viz., May 22.

The following has been disclosed by phenological observations. Winter oat and barley came at once into stalk in all variants August 28, and passed further stages of development simultaneously. Control plants of winter vetch started budding July 14 and flowering July 20. In plants yarovized ordinarily budding set in July 1; flowering July 5—7, i. e. two weeks earlier than in controls. The plants yarovized in water showed practically no

difference from controls, since they began to produce buds July 13—16 and to form flowers July 17—21.

Consequently, the usual yarovization procedure applied to seed of winter vetch has accelerated flowering for two weeks, while no acceleration whatever has resulted from the immersion of seed in water.

Unyarovized lupine plants commenced budding September 7; those yarovized ordinarily, August 23—25, i. e. nearly two weeks earlier. Budding was not so strongly accelerated in plants yarovized in water where it set in July 1, i. e. 5—8 days later than in ordinarily yarovized plants (Fig. 3).

If the seed of winter wheat Ukrainka, winter vetch or lupine is continuously kept in water at 3° C before planting, this fails to result in accelerated development of plants be it that they are spring or winter varieties. The reason lies in their delayed development during the first stage, which is passed normally by the plants at the same temperature when conditions of yarovization are available.

Fig. 3. *1*—grown from swollen seeds; *2*—ordinarily yarovized seeds; *3*—seeds kept in water for 20 days.

In spring wheat *Erythrospermum* 82/02, winter barley and oats the rate of their passing the first stage of development is neither enhanced nor reduced by water treatment of seed before planting. The length of the vegetation period is the same as in unyarovized plants. Nor can their development be accelerated by the ordinary method of yarovization. The evidence furnished by experimental data is that the plants which accelerate their development in response to yarovization carried on under usual conditions, nearly (Ukrainka yarovized during 40 days) or absolutely (winter vetch, lupine) refuse to do this after being treated by immersion in water.

In view of the fact that the respiratory function is very intensive in germinating seed and that the content of oxygen is rather poor in water, we may suppose that the delayed development of plants grown from immersion treated seeds is due to oxygen deficiency and its attendent derangement in metabolism.

Experiments on yarovization of seed in one medium have testified to the important rôle of oxygen in processes connected with transition of plants through the first stage of development. Corroborative evidence of this statement has been furnished by Purvis who was conducting the yarovization of spring wheat in an atmosphere, where the ratio of oxygen was only $^1/_{500}$ of its normal content in air. The development of plants thus treated coincided in time with that of plants from unyarovized seed.

On ground of the experimental data here referred to we are once more entitled to say that not until the requisite factors have combined in a definite complex, is the plant sure to pass any of its developmental stages.

Gasperov's one-sided interpretation of the procedure known as «cold germination» of winter plants is therefore obviously wrong. Positive influence of cold germination upon both winter and spring varieties can become apparent only in combination with other factors determining the transition through yarovization stage.

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ZOOLOGY

CONTRIBUTION TO THE COMPARATIVE ANATOMY OF THE GENUS *MURAENOLEPIS* (*GADIFORMES*)

By A. N. SVETOVIDOV

(Communicated by S. A. Zernov, Member of the Academy, 11.III.1939)

The genus *Muraenolepis* Günther 1880(1) which was known to Günther only from two specimens, was classed by him among the *Gadidae*. However, in a number of important anatomical characters (the gill-clefts are limited from above, commencing below the level of the pectorals, the scales are oblong and arranged at right angle to one another) *Muraenolepis* considerably differs from the *Gadidae*. Therefore Regan(2), who has pointed out the increased number of the pectoral radials as an outstanding anatomical feature of *Muraenolepis*, gave this genus the rank of an independent family of the order *Gadiformes* (*Anacanthini*). The pneumatic duct presumed by Günther has been supposed by Regan (l. c., p. 464) to be «probably a vascular and nervous strand supplying the muscular portion» of the air-bladder.

Goodrich(3) in 1909 pointed to a peculiarity in the structure of the outer orbital region and of the anterior part of the brain of *Gadiformes*. The brain-cavity of *Gadiformes* extends forward above the interorbital septum as a narrow channel; the olfactory bulbs are situated in the anterior part of this channel close to the nasal sacs and are connected with the forebrain by means of long olfactory tracts [details see in the previous paper by the same author(4)*].

Thus, some important features in the structure of *Muraenolepis* still remain unstudied. The anatomy of the specimen of *Muraenolepis microps* Lönnberg, kindly sent by Dr. Norman(5) of the British Museum, has been studied by me. The study of this specimen shows:

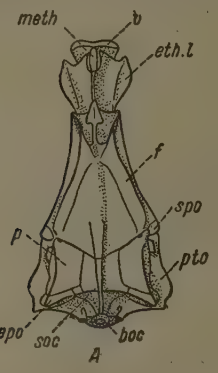
1. In full conformity with the presumption made by Regan, there is no pneumatic duct. The presumptive pneumatic duct is a mesenterial strand stretching from the anterior part of the air-bladder to the stomach; this strand is divided into two parts, one of which is fastened to the middle and the other to the hind part of the stomach. The air-bladder is small, very thick walled, without cornu-like outgrowths, arranged in the posterior part of the body-cavity beginning approximately from its middle. Pyloric appendages absent.

2. Olfactory bulbs are close to the nasal capsules, but not to the fore-brain. They are connected with the nasal capsules through openings in the

* Fig. 1. on page 1282 of this paper must not be taken into consideration, because of erroneous lettering.

lateral ethmoids by means of very short olfactory nerves and with the forebrain by means of long olfactory tracts. The olfactory tracts run close to each other all along in a special channel above the interorbital septum. The lower part of this channel above the interorbital septum and interorbital septum are membranous.

3. As in *Gadus*, opisthoticum exceptionally well developed, pierced by an opening for the glossopharyngeal nerve, partly bordering by its posterior ridge the opening of the n. vagus; is in contact with the basioccipital below it, thus separating the prootic from the lateral occipital. Supraoccipital is well developed, with a small longitudinal crest throughout its length. Frontals fused into



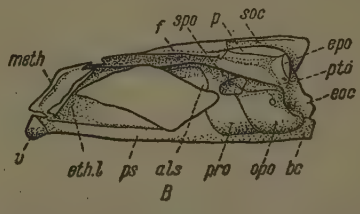


Fig. 1. Skull of *Muraenolepis microps* Lönnberg. *A*—from above, *B*—from the side, × 2. *als*—alisphenoideum, *boc*—basioccipitale, *eoc*—occipitale laterale, *epo*—epioticum, *eth. l*—ethmoidale laterale, *f*—frontale, *meth*—mesethmoideum, *opo*—opisthoticum, *p*—parietale, *pro*—prooticum, *ps*—parasphenoideum, *soc*—supraoccipitale, *v*—vomer.

an individual plate. The crests above the sensory channels on the frontal and the pterotic well developed. Orbitorostral part of the skull longer than the posterior one. Neurocranium widely opens in front, without fontanelles in the lateral occipitals. Basioccipital without tubercules below for the attachment of the muscles. Head of vomer toothless. Similar to *Gadiformes* in the structure of the skull and in other respects (Fig. 1).

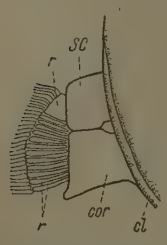


Fig. 2. Right pectoral girdle of *Muraenolepis microps* Lönnberg seen from outside, × 3½. *cl*—cleithrum, *cor*—coracoideum, *r*—radialia, *sc*—scapula.

4. The structure of the pectoral girdle in *Muraenolepis* is very peculiar. The number of the pectoral radials in the examined specimen is 13, and no less than 10, according to Regan. All the pectoral radials, except the upper one which is enlarged and short, are long and narrow (Fig. 2). The scapular foramen is between scapula and coracoid. In other respects is similar to *Gadiformes*.

Thus, *Muraenolepis* does not differ from *Gadiformes* in the structure of the forebrain and of the skull, differing essentially in the increased number of the pectoral radials. The increased number of the pectoral radials is of great importance and justifies *Muraenolepis* to be ranked as a suborder of *Gadiformes*. Thus, the order *Gadiformes*, on the basis of the present and previous investigations of the author[4] should be divided as follows:

Suborder 1) *Muraenolepoidei* (the genus *Muraenolepis* with three marine species in the southern hemisphere) with one family, the definition of which is given above;

Suborder 2) *Gadoidei* with the families *Moridae*, *Bregmacerotidae* and *Gadidae*, the definition of which is given in the above-mentioned papers

of the author. To the definition of the *Muraenolepidae* it must be added that there is no distinct caudal fin, the first dorsal consists only of one ray, gill-openings are restricted from above, commencing below the level of the pectorals, the scales are oblong and arranged at a right angle to each other. The *Muraenolepidae* are the most generalized family of all *Gadiformes*. In the structure of the skull they remind some of the *Gadidae* from the most generalized subfamily *Lotinae*, however, contrary to Regan's assumption, not *Onos* (*-Enchelyopus*), but the genera *Phycis* and *Urophycis*. The increased number of pectoral radials should be considered as indicative of generalization.

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ZOOLOGIE

RELICTOGOBIUS KRYZHANOVSKII n. g. n. sp., UN NOUVEAU GOBIIDÉ PROVENANT D'UN LAC SALÉ DE LA PRESQU'ÎLE ABRAOU (BASSIN DE LA MER NOIRE)

Par Z. M. PTCHELINA

(Présenté par S. A. Zernov, de l'Académie, 4.III.1939)

Pendant l'été 1934, S. G. Kryzhanovski trouva dans un petit lac salé de la péninsule Abraou, situé entre les crevasses Novo-Aguirskaya et Lobanova, quelques exemplaires d'un gobiidé qui se distinguait des gobiidés ordinaires de nos eaux par sa coloration charactéristique. En 1935 Kryzhanovski fit quelques nouvelles captures et obtient en tout 24 exemplaires de ce gobiidé dont la longueur variait de 28.3 à 65.5 mm et qu'il mit aimablement à notre disposition pour un examen et une comparaison aves les espèces connues.

Outre sa coloration, ce gobiidé se distingue par les caractères suivants: la présence d'une vessie natatoire, la présence exclusive d'écailles cycloïdales, un nombre restreint de rayons dans la seconde nageoire dorsale. La disposition des génipores est semblable à celle des gobiidés du genre *Gobius*, mais les canaux muqueux et les pores sont différents. Notre gobiidé n'a pas de canal muqueux au-dessus de l'operculum, les canaux muqueux antérieurs sont raccourcis, les pores σ s'ouvrent au bord antérieur de l'oeil, le pore λ se trouve près du milieu de la ligne que réunit les centres des pupilles.

Tous ces indices, ainsi que plusieurs autres, assez caractéristiques, nous permettent de rapporter ce gobiidé non seulement à une espèce nouvelle, mais à un genre nouveau—apparemment une forme relicte. Nous nommons cette espèce de gobiidés en l'honneur de S. G. Kryzhanovksi à qui appartient la priorité de leur découverte.

Relictogobius n. g. Corps allongé, comprimé dans la partie postérieure. Vessie natatoire présente. Tête aplatie. Toute la tête, une partie du dos, les bases des pectorales et le devant de la gorge sont nus (sans écailles). Le reste du corps est couvert de menues écailles cycloïdales. Séries transversales d'écailles 60—80. Bouche moyenne, les coins ne dépassent pas la verticale du bord postérieur de l'oeil. Narines étirées en tubes saillants. Langue arrondie. Plusieurs rangées de dents aux machoires. Canines. Pas de dents sur le vomer. Les muscles dorsaux recouvrent la boîte du crâne jusqu'aux yeux. Isthmus large. Canaux muqueux antérieurs raccourcis, les pores σ s'ouvrent près du bord antérieur de l'oeil. Point de rangée sous-orbitale longitudinale arquée de génipores. La nageoire caudale arrondie. La dorsale et l'anale courtes.

Relictogobius kryzhanovskii n. sp. Tête aplatie dont la hauteur correspond à 3/4 de la largeur. La longueur de la tête est comprise 3.8 à 4.4 fois dans la longueur totale et 3.3 à 3.7 fois dans la longueur du corps (sans *c*). La largeur de la tête vaut 65 à 80% de sa longueur chez les adultes et 59% chez les jeunes. Les angles de la bouche se trouvent sur la verticale du bord antérieur de l'oeil. La mandibule dépasse légèrement la mâchoire supérieure. La lèvre supérieure n'est point élargie latéralement. La longueur du museau et le diamètre de l'oeil sont presque égaux et correspondent séparément à $^1/_4$—$^1/_5$ de la longueur de la tête. L'espace interorbital est étroit chez les adultes et plus large chez les jeunes. Dans le premier cas il est compris 3 à 4 fois dans le diamètre de l'oeil, et dans le second—2.3 à 2.5 fois. La hauteur maximum du corps est comprise 5.2 à 6.8 fois dans la longueur du corps (moins *c*). Le plus souvent elle surpasse légèrement la largeur maximum du corps, mais peut lui être égale et même moindre. La longueur du tronc caudal surpasse de 1.5 à 1.8 fois sa hauteur. La ventouse n'atteint pas l'orifice anal et vaut 16 à 20% de la longueur du corps; la distance entre le sommet de ses rayons et l'orifice anal vaut 8.4 à 12.8% de la longueur du corps; la distance entre le sommet de ses rayons et l'orifice anal vaut 8.4 á 12.8% de la longueur du corps. Les palettes de la ventouse sont à peine visibles. Les rayons supérieurs des pectorales ne sont pas en forme de crin.

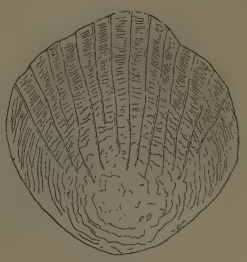

Fig. 1.

I *D* VI (VII), II *D* 1(9) 10—11*, *A* 1(8) 9—10.

Chez quelques exemplaires la seconde nageoire dorsale est d'une hauteur à peu près uniforme, tandis que chez d'autres elle s'élève vers le centre ou à l'arrière. L'anale s'élève à l'arrière. La tête, une partie du dos jusqu'à

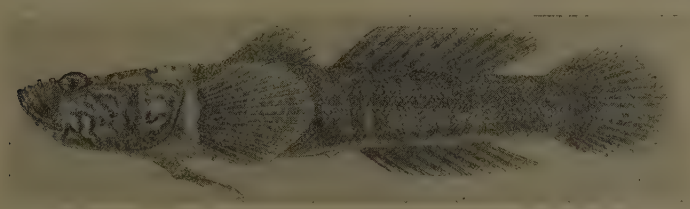

Fig. 2. Vu de côté. ×[1 1/2

la base des pectorales, les bases des dorsales et le devant de la gorge sont nus. Le reste du corps est couvert d'écailles exclusivement cycloïdales (fig. 1). Le nombre des séries transversales est 63 à 72.

Coloration: le corps est brun, le côté inférieur blanchâtre. La tête est couverte d'un dessin marbré caractéristique (fig. 2 et 3). Sur le corps se trouvent environ 10 raies foncées transversales et sur le dos 3 raies claires, dont l'une au commencement des pectorales, la seconde au commencement de la première dorsale et la troisième à la fin de la seconde dorsale. Toutes les nageoires sont foncées avec un étroit liséré clair. Les bases des péctorales sont claires presque sans pigment. La base des rayons des pectorales est bordée

* Les deux derniers rayons ramifiés de la dorsale et de l'anale qui se touchent par les bases ne sont pas considérés comme un seul rayon. Chez un exemplaire tous les rayons de la II dorsale et de l'anale étaient ramifiés.

par une raie noire recourbée. Les individus jeunes ont une coloration gris-jaunâtre.

Les canaux muqueux antérieurs sont raccourcis. Le pore λ se trouve environ au milieu de la ligne qui réunit les centres des pupilles; les pores σ s'ouvrent près du bord antérieur de l'oeil, un peu plus loin que la paire postérieure de narines. Fig. 4 et 5* montrent la disposition habituelle des canaux muqueux et des pores de ce gobiidé.

Fig. 3. Vu d'en haut. $\times 1^1/_2$.

Les génipores du devant de la tête sont plus gros et frangés. Les rangées *r* sont courbées, rapprochées vers les bouts antérieurs. Les rangées s_1 et s_2 sont le plus souvent arquées, s_3 en forme de petits tas allongés de hemicycle ou d'angle aboutissant à la lèvre supérieure.

Les rangées *c* sont habituellement recourbées formant un angle obtus dont le sommet est dirigé vers la lèvre supérieure. Les rangées *d* commencent par des points à peine visible partant de la 5-me rangée transversale sous-orbitale ou même encore plus loin et s'étendent jusqu'aux angles de la

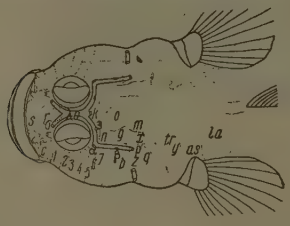

Fig. 4. Canaux muqueux, pores et génipores. Tête vue d'en haut.

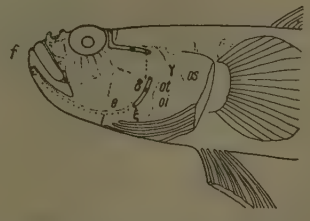

Fig. 5. Canaux muqueux, pores et génipores. Tête vue de côté.

bouche, où souvent ils se recourbent et se prolongent le long de la lèvre supérieure. Il peut y avoir des groupes de génipores entre les pores λ et x. Les rangées *f* sont en forme de petits tas allongés.

Le nombre de rangées transversales sous-orbitales est 6 à 7. Les deux ou trois premières rangées contiennent des génipores plus grandes. Les deux dernières sont séparées par une rangée transversale *b*, quelquefois mal exprimée, mais habituellement dépassant la dernière rangée transversale à une distance égalant l'intervalle entre 2 ou 3 rangées transversales. La dernière

* Nous avons découvert chez trois individus la présence de pores additionnels; chez l'un entre les pores α et ω, chez un autre entre λ et σ et chez le troisième entre β et ρ.

rangée transversale est le plus souvent interrompue et dépasse de beaucoup le niveau de la rangée *d*. La rangée *e* est parfois interrompue près des coins de la bouche. La rangée *i* est composée de génipores rares et gros.

Les rangées de la nuque sont bien exprimées chez certains gobiidés et à peine visibles chez d'autres. Elles partent du pore α et dépassent le plus souvent le pore ω. Les rangées *o* sont obliques et se rejoignent complètement ou presque complètement vers le milieu. Les rangées *g* et *m* ne sont pas toujours bien définies. La disposition des autres génipores varie, mais en général elle est pareille à celle des représentants du genre *Gobius*. Sur les flancs se trouvent 10 à 20 rangées transversales de génipores.

D'après les données de S. G. Kryzhanovski et de L. I. Volkov, le petit lac d'où proviennent ces gobiidés est situé entre les crevasses Novo-Aguirskaia et Lobanova et s'étend le long de la côte. Il est séparé de la mer par une jetée (environ de 50 m) de grosses pierres angulaires. Le lac n'a pas de communication avec la mer, mais il est possible qu'un échange d'eau ait lieu entre les pierres de la jetée. Le lac a une étendue de 22 712 m² et une profondeur de 2 à 3.5 m. La teneur de chlore est de 11.16%, ce qui correspond à une salinité d'à peu près $20^0/_{00}$. Près des rives éloignées de la mer croissent en grande quantité *Phragmites* et *Potamogeton*. En fait d'animaux le lac contient un nombre énorme d'*Upogebia*, de grosses actinies, des éponges, des crevettes et des isopodes. Nous avons trouvé des restes d'*Upogebia* dans le canal dijestif de notre gobiidé. Outre ce gobiidé qui s'y trouve en grand nombre nous ne découvrîmes dans le lac point d'autres poissons. Les gobiidés se tiennent près des rives pierreuses, sous les pierres et entre les touffes de *Potamogeton*.

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EXPERIMENTAL MORPHOLOGY

EXPERIMENTAL ANALYSIS OF DEVELOPMENT OF PLUMAGE COLORATION IN *PERDIX PERDIX* L.

By B. G. NOVIKOV

(Communicated by I. I. Schmalhausen, Member of the Academy, 5.IV,1939)

In a previous paper(1) it was stated that partial monomorphism for the horse-shoe shaped spot is due to lesser response of this part of plumage in the female partridge to the action of the sex-hormone. The amount of hormone present at the time of natural moulting is insufficient to inhibit the development of the brownish pigments, and the spot develops in the female too. In this connexion it seemed probable that the development of characters of complete monomorphism is due to a further decrease, and perhaps to a complete loss of response in feather-forming tissues of the female to the action of the ovarian hormone. The possibility suggested itself that the ovarian hormone present during the natural moulting period does not reach the threshold of response of the feather papillae which results in the development of the male-type plumage. If this assumption be correct, it might be expected that in the presence of higher concentrations of ovarian hormone the potency of developing male character would be suppressed, and one would succeed to induce the growth of henny-feathering absent in the normal bird. Such an experiment can be performed in two ways: either by way of artificial stimulation of growth of feathers in females during the period of maximum activity of their own ovary, or else by treating with a female sex-hormone preparation.

To check the above assumption, were carried out experiments on castration, production of new generations of feathers within periods of various gonad activity and sex-hormone injection. Observations on 35 females and 30 males were made during the period 1937—38. In all the operated and normal birds small areas of skin were deplumed every month in different parts of the body. The nature of pigmentation in the regenerated feathers was studied macroscopically.

As a result of these investigations was obtained a negative effect of gonad extirpation on the development of characters of monomorphism in the plumage. In all the operated birds, irrespective of the time of deplumation, feathers of new generations always displayed normal colouring. Upon removal of the ovary, merely the female wing coverts were observed to assume the male colouring (character of dimorphism). As opposed to ovariotomized birds, normal females failed to show changes in the pigmentation of regenerated feathers only during the period of low activity

of gonads (July-May). During the period of increased sexual activity (May-June), on the contrary, extremely sharp changes in the plumage colouring of all the females took place. Ash-grey, «rippled» feathers of the neck and crop were replaced by feathers of dark colour with light tubular and transverse stripes. The ash-grey «rippled» feathers of the sides with a broad ochraceous band assumed a dark brownish hue and light transverse stripes. The rust coloured feathers on the back with brownish transverse stripes were replaced by dark coloured feathers with a light transverse band across the middle. Only the ochraceous colour of feathers on the throat and neck did not change. Control autopsies of females of these series brought to show a sharp hypertrophy of ovaries and oviducts (separate follicles attained 8—10 mm in diameter, and in some instances ovulation was found). Thus, in the case of increased activity of the ovary, colouring in females looses to a certain extent its regional character giving place to a uniformly spotted primitive garb, almost identical with the juvenile one and the henny-feathering of allied dimorphous species. The second series of experiments involved injection of the female sex-hormone, an amount of 300 to 500 M. E. having been administered every day. Tests on males and castrated females were run mainly in the summer, on normal females during the period of non-functioning of gonads. Feathers developed under the action of the hormone injection showed exactly the same changes in pigmentation, as observed in females plucked during the period of high sexual activity. Both in castrated males and normal ones treated with female hormone, identical results were observed. Upon doses of 300 M. E. the effect produced in the latter was not complete; in such cases we succeeded but in inducing female coloration of the wing quills, inhibiting the development of the brownish horse-shoe mark and modifying the colouring of the feathers but in some pteryliae (the nape). Other experiments have likewise revealed differential response of some of the characters to the sex-hormone. Thus, by using the method of deplumation it was ascertained that signs of dimorphism in plumage develop normally even during the period of the lowest activity of the ovary (autumn-winter), whereas inhibition of the development of characters of partial monomorphism may be achieved ([1]) within March-July, and characters of complete monomorphism are obtained only during the period of maximum activity of the ovary (May-June). The above results point to the fact that feather papillae developing characters of constant dimorphism show the greatest susceptibility to the ovary hormone. On the other hand, tissues showing character of monomorphism are characterized by extremely feeble response. In this respect characters of partial monomorphism occupy an intermediate position. Besides, separate regions of the skin bearing characters of the latter two categories possess in their turn differential thresholds: feather papillae, developing male characters at a later stage of ontogeny, are more sensitive to the hormone. Thus, if on the basis of the studied characters one would try to trace the transition from dimorphism to monomorphism, such transition would be characterized by gradual decline of response of tissues to the ovarian hormone. Consequently, the process of transformation of sex characters into specific ones was effected in *Perdix perdix* at the expense of liberation of tissues from the determining influence of gonads.

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ANIMAL PHYSIOLOGY

ON THE PHYSIOLOGICAL PROPERTIES OF THE NERVELESS REGION OF THE SKELETAL MUSCLE

By I. BERITOV, Member of the Academy

Certain authors have found of late essential differences in the physiological properties of the nervous and nerveless regions. Thus e. g., the Treppe-phenomenon and the heightening of the excitability of the active muscle, the arising of bioelectrical currents of excitation or of the so-called currents of action, also the phenomenon of the pessimal effect, the sensitivity to acetylcholine are regarded as properties of the nervous region of the muscle ([1–6]). The ending of the motor nerve or the nerve plate in the muscular fibre is considered the morphological element, determining these phenomena.

On the other hand, we have shown that the nerveless region differs in nothing particular from the nervous regions. Both, if isolated, can be stimulated and respond by excitation and contraction, although these excitation and contraction are more intense in nervous regions than in nerveless ones ([8,9]); in connexion with the excitation of both arises the absolute refractory phase, however, in nervous regions it is shorter than in the nerveless ones ([10]); while the action currents arise in both, only they are stronger in the nervous regions ([8]), and the latent period of contraction is shorter than in the nerveless ones ([10]). The same difference was established by the same authors in curarized muscles as well.

In the present communication we report on the results of our recent studies concerning the physiological properties of the totally nerveless proximal region of m. sartorius, obtained with the aid of the method of myographic registration of mechanic effects.

Proximal regions of m. sartorius, 7—8 mm long, i. e. less than one fifth of the entire length of this muscle, usually contain no motor endings. We ascertained this by means of histological examinations of series of preparations. These nervous elements are usually located at a distance of 9—10 mm from the proximal end. Here passes one of the nervous zones ([7]). The physiological investigation of such proximal regions which are absolutely devoid of motor nerve elements, shows that these areas, while in a perfectly fresh condition, possess every property generally belonging to an entirely uninjured muscle.

If the muscle is stimulated by induction shocks of moderate intensity every 1—2 sec., the mechanical effect of the muscle is increased with every new shock. The typical Treppe-phenomenon is the result. After a

dozen of such stimulations the contractive ability of the muscle is heightened for a long interval, a minute or more. Thus in exp. *A* (Fig. 1) the mechanical effect was raised by 50% as the result of 12 stimulations. The experiments after 6 and 15 sec. showed a heightened effect. Similarly in exp. *B* which was performed 2 min. later, the first effect was higher than in the beginning of exp. *A*. Consequently, the increased ability of response was maintained during these two minutes. This heightening of the ability to respond is manifested with particular force in the case of tetanic stimulation. After a short (2 sec.) but strong electric stimulus the threshold force of an identical stimulation produces a much more intense contraction, than before the tetanic stimulation (Fig. 1, exp. *B*).

The heightening of the contractive ability takes place in the entire proximal area of the muscle. This may be seen from the following experiment. If the tetanic stimulation is applied at a distance of 1—4 mm from the proximal end and the stimulation by single induction shocks is applied at a distance of 5—8 mm from it, i. e. 2—4 mm from the tetanized site, the individual contractions after the tetanic stimulations are greatly increased. In some cases this heightening is manifested only during a few seconds, in certain cases it can be maintained for minutes. It seems that the fresher is the preparation, the higher is its functional condition, and the longer the heightened responsiveness of the muscle after the tetanic contraction.

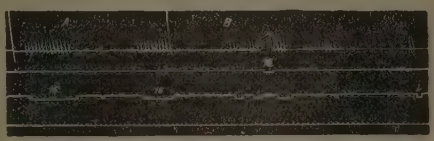

Fig. 1.

Fig. 1. 9. IV. 1938. M. sartorius of a newly caught frog. The total length of the muscle is 44 mm, the proximal region is taken 8 mm long. For tetanic stimulation the electrodes are placed at the distance of 1—4 mm. The threshold of stimulation is 22 cm distance from the induction coil.

In exp. *B* tetanic stimulations are performed at a 18 cm distance from the coil. The frequency of the stimulation is 30 per 1 sec. (upper signal). For provoking single contractions the electrodes are applied somewhat higher at the distance of 5—7 mm from the proximal end (lower signal). Below: the time expressed in seconds. Histological examination has shown that the area tested contains no motor nervous elements.

The phenomenon of the heightening of the contractive capacity in cut off proximal regions of the muscle in connexion with the active condition is observed almost in every preparation provided it is fresh, i. e. tested soon after the operation, is not injured by frequent irritations and is not taken from an exhausted frog. But every time after numerous stimulations in the course of one hour the muscle is brought into such a condition that individual induction shocks produce a weakening of the effect from the very beginning and do not produce any heightening even after a tetanic effect.

It is characteristic that even in the most fresh preparations it is possible to observe weakening instead of intensification, if only the tetanic stimulation is administered for a relatively long time, some 10 sec. or more. The excised bloodless proximal region is soon fatigued by tetanic stimulations, already after a few seconds. If the tetanic stimulation is stopped during the period when the curve of contraction is descending, a weakening of single contractions is regularly provoked.

It might be thought that the heightening of the functional activity of the proximal areas is determined by the electrotonic action of the stimulating

current itself and not by the active condition of the muscle. The following facts speak against this assumption. The described heightening of the functional condition is observed in the proximal area also in the case of its being brought into an active condition through the nerve. The entire muscle is taken, its proximal area is stimulated by isolated induction shocks and the influence of the tetanic contraction provoked by the stimulation of pl. ischiadicus is observed. As Fig. 2 shows, in the case of a weak tetanic contraction the single contractions remain unaltered, while in that of strong tetanus they are greatly intensified (exp. *B*). In this case as well the phenomenon is observed only in fresh preparations. After a series of stimulations a similar tetanic stimulation does not intensify, but to the contrary weakens the single contractions (Fig. 2, exp. *C*).

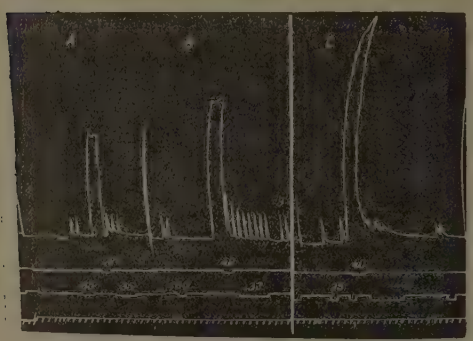

Fig. 2.

Fig. 2. 3. III. 1938. The entire m. sartorius is recorded. The peripheral end of the dissected plexus ishiadicus is subjected to tetanic stimulation at 46 and 40 cm distance from the coil. The proximal area at the distance of 2—5 mm from its end is stimulated by isolated induction shocks. Exp. *A* and *B* were performed when the preparation was fresh, exp. *C* after it had been fatigued. The sweep of the myograph in the last exp. was doubled, which explains the greater amplitude of the curve of tetanic contraction. Below—time in seconds.

It must be noted that in fresh muscle the thresholds of stimulation of its proximal nerveless areas are relatively low: 25—35 cm distance from the coil, while the threshold of the plexus ischiadicus is 45—50 cm. It might be suspected that contraction after the stimulation of the nerveless proximal areas is also effected by the action of the loops of the stimulating current on nervous elements. Experiments, however, contradict such an assumption. Owing to the relatively high conductivity of muscular tissue, the loops of the current are insignificant even at a distance of a few millimeters from the electrodes. This can be easily verified by means of a spring galvanometer. Yet another simpler proof is available. If we first determine the thresholds of the proximal area in the nerveless area of an entire muscle at 2—4 mm distance from its proximal end and afterwards separate this nerveless area from the rest of the muscle by cutting it off, the thresholds of stimulation of this area remain unchanged. This means that already the threshold force of electric stimulation produces its effect on the proximal area of the entire muscle by means of a direct action of the current on the muscular excitable system.

Thus, both the entire normal muscle in a fresh condition and its totally nerveless area manifest a heightened responsiveness after tetanic or isolated contraction.

The proximal nerveless area of m. sartorius shows a pessimal and an optimal effect also in respect of the frequency of stimulation. At a certain high frequency of stimulation—100—250 per 1 sec.—the mechanical effect of the muscle shows a typical pessimal character. A quick beginning bearing the character of a shudder is followed by a more or less rapid drop (Fig. 4).

On the other hand, at a certain low frequency—30—40 per 1 sec.—the shortening of the muscle achieved by means of a quick contraction holds on for many seconds or somewhat weakens after the initial indent and then slowly increases again (Fig. 1), or the contraction intensifies gradually from the very beginning (Fig. 3).

Usually the pessimal and optimal phenomena depending on the frequency of stimulation are studied by means of continuously changing the frequency of stimulation from the lowest one to the highest or vice versa. On the proximal nerveless area it is also easy to observe that every time the frequency si raised above a certain level, the mechanical effect experiences a fall. Thus e. g., in Fig. 3 the tetanic stimulation with the frequency of 40 shocks per 1 sec. is several times alternated by one with a frequency of 70 shocks

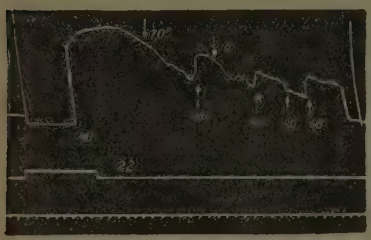

Fig. 3.

Fig. 4.

per 1 sec. and every time the rise in the frequency is accompanied by a speediest fall of the curve of contraction, while the return to the lower frequency leads to a very quick rise of this curve.

Fig. 3. 13. IV. 1938. Proximal area, 8 mm. Stimulated tetanically at 2—4 mm from proximal end. Power of stimulation: 22 cm distance from coil (threshold—27 cm). Frequency of stimulation 40 per 1 sec., thrice during the experiment this frequency is alternated for a short time by a frequency of 60—80 per 1 sec. Below—time in seconds. Histological examination revealed no motor nervous elements.

We tried many times to obtain the phenomena of optimum and pessimum dependent on the intensity of stimulation at frequencies varying between 40—250 per 1 sec. Yet in no test did we succeed in obtaining a sinking of the mechanical effect in connexion with increased intensity of stimulation. Just the opposite was the case: every time each new intensification of the stimulation provoked a fresh increase of the mechanical effect.

The cause of this is, in the first place, that every intensification of the stimulation involves into the contraction new less excitable muscular fibres, and in the second place the process of contraction extends to an ever increasing length of the muscular fibres owing to an intensified progression of the loops of the current. By this reason the intensification of the faradic current leads to the intensification of the mechanical effect.

It is characteristic that the heightening and lowering of single contraction may be likewise observed after a pessimal contraction. If this pessimal contraction continues only a few seconds, its effect on the single contractions is favourable, the heightened responsiveness holding on for dozens of seconds. If, on the other hand, the pessimal contraction continues for many seconds, then, contrariwise, the single contractions provoked after this prolonged pessimal condition are greatly weakened. They may afterwards gradually become stronger and, finally, after many seconds rise above the level, which they attained before the pessimal stimulation (Fig. 4).

Fig. 4. 14.IV.1938. Proximal area, 8—9 mm. Stimulated tetanically at a frequency of 100 per 1 sec. at 2—4 mm from the proximal end (distance from the coil 20—17 cm) and by isolated shocks at a distance of 6—8 mm (coil distance 30 cm). A short pessimal stimulation raises the subsequent single contractions, while a lengthy one, oppositely, weakens them. Histological examination revealed 2 nerve bundles 8—9 mm from the proximal end. The rest of the muscular fragment is free from motor nervous fibres.

It follows that the process of stimulation in the nerveless region is accompanied by refractory phases, and therefore it can be affirmed that the nerveless area submits to the law of excitation, the so-called «all or none» law. Moreover, the same physico-chemical processes take place in the nerveless region during both pessimal and optimal contractions. In both cases they not only determined contraction, but alter for a long period the excitability of the muscle, first raising and then lowering it.

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ANIMAL PHYSIOLOGY

ON THE ORIGIN OF THE ACETYLCHOLINE CONTRACTURE

By I. BERITOV, Member of the Academy

It is known that acetylcholine provokes in the skeletal muscle both a quick contraction determined by excitation and a contracture without excitation. We studied in detail the character of the effect of acetylcholine on m. sartorius and m. pectoralis p. abd. of a frog and found that there is a definite difference in the action of acetylcholine on either muscle. Neither in muscles with intact circulation nor in isolated (cut out) muscles does acetylcholine always provoke both types of contraction. Quick contraction, recorded by the myograph or visible to the eye as a twitching of individual bundles of fibres, takes place only if the functional condition of the preparation is good. In cut out muscles, accordingly, since their functional condition is evidently abnormal, quick contraction is observed, but very seldom in the case of m. pectoralis p. abd., while in the case of m. sartorius it is frequently absent. In these cases the contraction bears from the very beginning the character of a contracture (Fig. 1).

Quick contraction is absent or greatly weakened as the result of injury in connexion with the cutting out of the muscle, as the result of blood-draining, of fatigue, of repeated acetylcholine poisoning. Quick contractions are also absent in muscles obtained from exhausted frogs, even if these muscles are not excised and the circulation in them is preserved. In all these cases the excitability of the muscles is relatively low, but their contractibility in response to a strong stimulus, direct or indirect, may be high—over 50%. Also the contracture provoked in such muscles may be high, particularly in m. pectoralis p. abd., which may show in the absence of twitch a contracture up to 50% of its normal length (Fig. 1).

It is characteristic that out of over 100 tests, in which concentrations of acetylcholine varying from 1 : 1 000 000 to 1 : 500 were applied to cut out m. pectoralis p. abd., a clearly marked contraction was observed but several times. At the same time in m. sartorius it was obtained more or less regularly and only rare preparations from emaciated frogs did not produce it. In numerous cases we cut out both muscles from the same frog, connected them immediately with the myograph and subjected them at once one after the other to the action of acetylcholine. Usually the result obtained was such as is presented in Fig. 1. M. sartorius responds to acetylcholine first by a speedy contraction and later by a contracture, while m. pect. p. abd. responds by a contracture from the very beginning.

Fig. 1. 8.IV.1938. Records of m. pect. p. abd. (above) and m. sartorius (below). Both muscles are from the same frog. Exp. *A* shows the myograms of a tetanic stimulation of m. sartorius. The maximum contraction of the

muscle was 12 mm. Exp. *C* shows its contraction after the application of acetylcholine (diluted 1 : 20 000, several drops of the solution are added). In this test the muscle produces first a series of twitches and then a contracture which after 6—7 sec. attains its maximum, equalizing a contraction of 7 mm, i. e. about 18% of the muscle's normal length. Exp. *B* shows the tetanic effects in m. pect. p. abd. Maximum shortening—22 mm. In exp. *C* the same muscle was poisoned with acetylcholine 1 : 20 000. The contraction increases slowly during one minute and reaches 21 mm, or 50% of the muscle's normal length. At the moment marked by an asterisk, the myograph was incidentally jolted and the muscle was somewhat distended. The moments of the stoppage of the kymograph are indicated by arrows. The figures on the signal line designate the distance of the induction coils in cm. Below—the time in seconds.

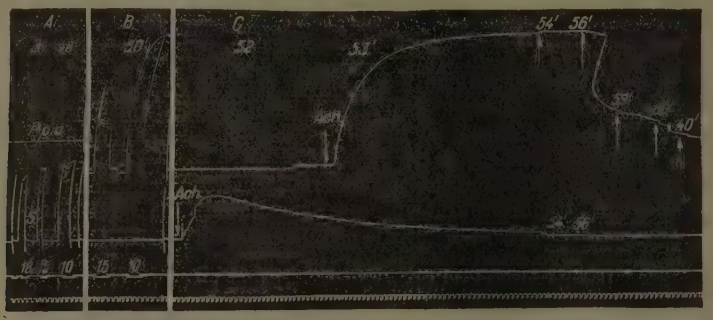

Fig. 1.

The character of the contracture effect in m. pect. p. abd. is always like in Fig. 1 : a slow gradual rise during several dozens of seconds, then a protracted plateau for several minutes and, finally, an extremely slow sinking during many minutes (5—10 and even more). M. pect. p. abd. shows typical differences from m. sartorius in respect of contracture. In the latter muscle the rise of the contracture lasts only several seconds, and its maximum being reached, the contracture begins immediately to relax; the entire effect holds on not more than 1—1½ min. (Fig. 1).

It is characteristic, moreover, that m. pect. p. abd. always produces a more considerable contracture shortening than m. sartorius. The first muscle in extreme cases is shortened by 50% like in tetanus. The shortening of the second after acetylcholine poisoning never reached 50%, though after tetanic stimulation this muscle also may become shorter by 50—60%.

Thus, the attitude of m. sart. and m. pect . p. abd. to acetylcholine is not the same. Each of the two muscles responds to this stimulus in its own particular way: m. sartorius in the absence of circulation performs an initial quick contraction regularly, while m. pect. p. abd. does so but occasionally; in general, m. sartorius performs a much less prolonged and weaker contracture, than m. pect. p. abd.

In muscles which have not been cut out and the circulation of which has remained undisturbed, if moreover they are tested absolutely fresh, acetylcholine produces every time a quick contraction together with a contracture. Yet, here also like in excised muscles, the contracture in m. pect. p. abd. increases much more slowly and continues for a much longer time than in m. sart. In these experiments only about 1 cm of the distal end or beyond the proximal end of the muscle was prepared for connecting it with

the myograph, and the action of acetylcholine was tested either by pouring an acetylcholine solution or applying lignine soaked in this solution along the muscle's length (Fig. 2).

Fig. 2. 10.I.1939. M. pectoralis pars abdominalis, intact. The recording is done from the area beyond the proximal end. In exp. *A* a drop of acetylcholine diluted 1: 10 000 is poured on the proximal end of the muscle. The muscle is inclined with the proximal end down. Acetylcholine has covered an area not exceeding 10 mm from the proximal end. The result is at first a quick contraction and then a slowly increasing contracture. In exp. *B* a drop of the same solution is applied to the middle of the muscle: an intense and quick contraction in the beginning, then a slow increase indicating the development of a contracture. In exp. *C* a tetanic contraction provoked by electric stimulation of the muscle. Time is expressed in seconds.

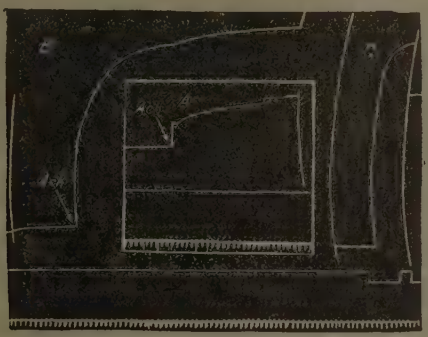

Fig. 2.

Tests with the local poisoning of small areas showed that this double effect is obtained both in nervous and nerveless areas of the muscle. As nerveless areas we used the proximal end, 8—9 mm long, of m. sartorius and the distal end, 7—8 mm long, of m. pect. p. abd. (6). Acetylcholine was applied at a distance of 2—4 mm from the end. The contraction started immediately and, if the muscle was in a good functional condition, its course was quick and followed by a contracture; if, on the other hand, its condition was bad, there was only a contracture. But of course, both the quick contraction and the contracture were considerably weaker in nerveless areas than in those with nerves, similarly to what was observed in respect of the effect of electric stimulation (cf. preceding report).

These results give us a new idea of the attitude of skeletal muscle towards acetylcholine. From the work of Sommerkamp (1) and Wachholder (2) we know that in this respect the striated muscles can be divided into 3 groups: one responds by a quick contraction, the other by a contracture only, the third by contractions of both types together. These authors place m. sartorius in the first group and m. pect. p. abd. in the second one. Our experiments show that both muscles in case their functional condition is good and more or less normal perform under the action of acetylcholine in their innervated and nerveless areas like—both twitches and contractures. The difference between these two muscles concerns only the character of the contracture, as this has been in detail described above. Consequently, the division of the muscles in groups according to their attitude towards acetylcholine needs correction. In dividing them thus, one should

consider only the character of the contracture and in this regard the skeletal muscles should be divided into two groups only: the group of muscles with a short contracture, like m. sartorius, and the group of muscles with a prolonged contracture, like m. pect. p. abd.

Moreover, our experiments show that, contrary to the affirmations of Rosenblueth and Luco (7), Brown (3), Riabinovskaja (8) et al., acetylcholine is not a specific stimulator of the nervous area, but affects the excitable system of the muscle generally.

There can be no doubt that acetylcholine provokes quick tetanic contractions via the excitable system. This is indicated in the first place by the circumstance that this contraction is propagated along the entire muscle, is accompanied by action currents [(3,4) et al.]. It follows that acetylcholine like many other chemical substances stimulates the excitable system. Whether a muscle responds to acetylcholine by a stronger or weaker tetanic contraction or does not respond at all, the underlying cause of this is the degree of excitability of the muscle. This explains why an injured, exhausted or fatigued muscle does not respond to acetylcholine by a twitch. Evidently, this is determined by the low excitability of such muscles, by the low level of the excitable system, due to which they become unable to be excited by acetylcholine.

The same cause is responsible for the fact that cut out m. pect. p. abd. show but rarely tetanic contractions, while cut out m. sart. perform such contractions with great regularity. It appears that the excitability of the first muscle is strongly lowered in connexion with its excision and loss of circulation. In the tests of L. Tskipuridze (a member of our laboratory staff), the chronaxie of an excised m. pect. p. abd. is almost 3 times that of m. sart., both in their innervated and nerveless areas. At the same time if the muscles are not cut out and continue to be supplied with blood (preparation with destroyed spinal cord), the chronaxie of m. pect. p. abd. is but little above that of m. sart. On the ground of these results there can be no doubt that the absence of quick tetanic contractions in the excised m. pect. p. abd. is determined by the low excitability of the muscular fibres.

Acetylcholine contracture shows characteristic distinctions from the twitches. It is limited to the poisoned area (4).

In spite of such a difference in the chief characteristics of the effects, experimental analysis shows that acetylcholine contracture takes place via the excitable system. This is to be seen first of all from that the lower the excitability of a muscle or of a muscle area, the weaker is the contracture and the higher must be the concentration of acetylcholine to provoke a contracture. Further, the fact that not only during tetanic contraction, but also during contracture the excitability and conductivity of the muscle sink, is another argument in favour of the same. Apparently, in the course of a contracture, the excitable system is discharged and split, like in tetanic contractions. This is indicated, on one hand, by the observations of Hess (5) that the fatigued muscle loses equally its ability to tetanus and to contracture, and on the other hand, by the observations of Wachholder (2) that when maximum tetanus is provoked by other means acetylcholine is unable to raise it.

We also arranged experiments for the elucidation of the rôle of fatigue in acetylcholine contracture. We fatigued the muscle by means of prolonged tetanic stimulation of the nerve or of the muscle itself and afterwards tested the action of acetylcholine. It was found that the stronger is the fatigue, the less is the effect of acetylcholine, i. e. the weaker the contracture provoked by it, and when a definite high degree of fatigue is reached, acetylcholine produces either no effect or a quite insignificant one. It clearly fol-

lows from these experiments that both tetanus and contracture are determined by the activity of the excitable system.

We arranged one more series of experiments with the lowering of excitability by means of ether narcosis. We found that in these experiments the action of acetylcholine on the muscle decreases parallel with the fall of excitability response to electric stimulation, and that in deep narcosis, when direct tetanic stimulation ceases to produce more or less considerable contraction, acetylcholine ceases to provoke any contraction whatever. If after it had been subjected to narcotization, the muscle is placed into Ringer-Locke solution for 10—30 min. and then again tested by electric stimulation and by acetylcholine, it is found that the ability of the muscle to respond by contracture to acetylcholine is restored simultaneously with its ability to be excited by electric stimulation.

Thus, our experiments leave no doubt that acetylcholine contracture takes place through the excitable system.

We think that acetylcholine in subminimum, subthreshold concentrations produces a partial splitting of the excitable system, this splitting being localized in the area on which the poison acts. This partial splitting intensifies the chemical processes which lead to contraction. Therefore a contraction arises in the poisoned area ([4]).

But this partial splitting occurs in so insignificant a measure that it is unable to provoke such an oscillation of the electric potential or such a bioelectric current which would determine a propagation of the process of splitting of the excitable system and consequently of the process of contraction. After this partial splitting the excitable system is restored. Consequently, until there remains some acetylcholine within the muscle fibres, the excitable system splits again and again, maintaining the already started local contraction.

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ANIMAL EMBRYOLOGY

INFLUENCE OF LOW TEMPERATURE ON DIFFERENT STAGES OF DEVELOPMENT OF EGGS OF BREAM (*ABRAMIS BRAMA*)

By T. I. PRIVOLNEV and A. M. RASUMOWSKY

(Communicated by S. A. Zernov, Member of the Academy, 11.III.1939)

Influence of low temperature upon the embryonic development of cold-blooded animals and particularly of fishes has not yet been studied sufficiently. The works by Loeb (2), Kellicot (1) and Stockard (5) should be mentioned; the material studied by the above-mentioned authors consisted in developing eggs of a small fish, *Fundulus heteroclitis*.

Kellicot has noted that this is the initial stage of development of the eggs of *Fundulus* which is distinguished by increased sensitivity to low temperatures. Stockard points to that stages of the appearance of many organs of the embryo, periods of differentiation, at which a temporary and extremely high activity of organs is observed, are periods of high sensitivity.

As may be seen from the data referred above, the question on the critical periods of embryonic development under the influence of low temperature requires further study. Study, of the influence of low temperature upon different stages of embryonic development of fishes, moreover, is of indoubted interest both to development mechanics and to practice. Eggs of fish often have to be transported for long distances, when ichtyofauna of new reservoirs is formed, or when most valuable species of fish are introduced to new regions of this country. To prevent hatching, eggs of our spring-spawning fishes are usually kept at low temperature which arrests their development; it is quite natural that a study of this problem is urgently needed.

The object of the present investigation was the study of the resistance of bream eggs to the action of low temperature at various stages of embryonic development.

Mature eggs of bream *(Abramis brama)* were fertilized by the «dry method», i. e. eggs obtained from the female were mixed with sperma without adding water. After 5—10 min. water was added, eggs were washed from the excess of sperma and placed into glass vessels where they were allowed to develop further. At various stages of development, beginning by fertilization and almost to hatching, eggs were placed into refrigerators at temperatures of 0, 3 and 6°C for 12 and 24 hours. After eggs had remained in refrigerators during the above time, they were placed again into normal temperature conditions (16—18°C), the proportion of dead eggs was determined and observations were conducted on embryos which continued development after treatment with low temperature.

Influence of Low Temperature during 12 Hours. The results obtained by low temperature treatment are presented in Table 1 and Fig. 1.

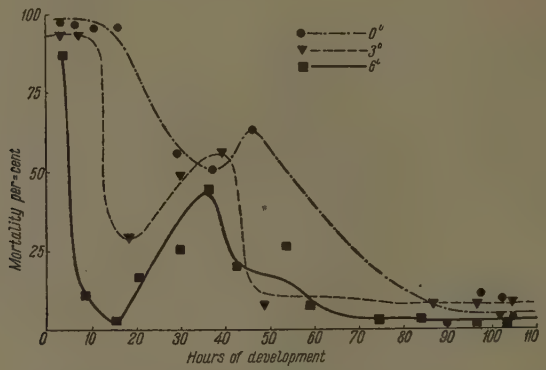
Table 1

Experiment №	Hours following fertilization	Developmental stages	Died after 12 hour treatment, %			Notes
			0°	3°	6°	
1	4	2—4 blastomeres	100	100	85	Further development results in monstrosity
2	8	Morula consisting of large cells	100	—	11	
5	12	Beginning of gastrulation	100	100	—	
6	16	1/3 of the yolk is grown over	100	—	4	
6a	20	1/2 of the yolk is grown over	100	30	18	
7	30	Prior to closing of the blastopore	59	50	26	The embryos formed after the influence of 0° and 3° C are largely monstrous
8	38	Closing of the blastopore	54	—	48	
9	42	Formation of the embryo	59	59	21	
11	50	Noticeable eye-vesicles	65	8	—	
12	55	Appearance of the lens	—	—	26	
13	60	Formed embryo	—	—	10	
14	75	» »	—	—	4	
15	82	Appearance of blood circulation	—	—	5	
16	86	—	—	10	—	
17	90	—	2	—	—	
17a	96	Prior to hatching	14	10	2	
18	103	» » »	12	6	—	
18a	105	Hatching	6	11	0	

As may be seen from Table 1 and Fig. 1, the eggs possess great sensitivity at the beginning of development, from fertilization up to the stage of large-celled morula. The proportion of eggs dying at this period of development under the influence of low temperatures reaches 100%. However, the result obtained at the temperature of 6°C stands somewhat apart. Here a small proportion of eggs usually remains alive and continues further development upon being placed into normal temperature conditions; however, development is abnormal, either not reaching embryo formation or a monstrous unviable embryo being formed.

When treatment is applied at a later stage of development, from large-celled morula to embryo formation, the proportion of dying eggs is greatly reduced. The eggs become more resistant. Then, beginning by the stage of initial embryo formation prior to closing of the blastopore up to the occurrence of the lens, period of the most active differentiation, the proportion of dying eggs under the same influence strongly increases again; the figure shows it most clearly for the temperatures of 3° and 6°C. The proportion of dying eggs fluctuates about 50%. Eggs which survived treatment, as a rule, produce further monstrous embryos. Monstrosities are of most varied kind: underdeveloped head, underdevelopment or absence of eyes, abnormally developed heart, distorted spinal column etc. The development

suffers strongest aberrations after the influence of a temperature of 0°C. If treatment is applied at later stages, when the embryo is already formed, the heart begins to function up to hatching of the embryos. The proportion of embryos succumbing to the same influence is sharply lowered. Data obtained from treatment with the temperature of 3° and 6°C agree with each other, while those obtained from treatment with the temperature of 0°C stand somewhat apart. The temperature of 0°C evidently produces a too strong effect, so that no periods of different sensitivity can be distinguished in early stages when eggs are most susceptible.



Influence of Low Temperature during 24 Hours. The results obtained from treatment with low temperature during 24 hours are analogous to those presented by Table 1 with the only difference that periods of different sensitivity are not clearly enough expressed up to the stage of formed embryo.

Table 2

Experiment №	Hours following fertilization	Stages of development	Mortality % after 24 hour treatment			Notes
			0°	3°	6°	
1	1	Prior to cleavage	100	100	100	
5a	2	Beginning of the cleavage	100	100	100	
6b	4	2—4 blastomeres	100	100	85	
7a	6	16—32 blastomeres	95	70	42	In further development monstrous embryos are formed
8a	8	Large-celled morula	100	65	40	
8b	10	Medium-celled morula	100	71	16	
10	13	Beginning of gastrulation	100	70	50	
10a	14	Do	100	80	—	
13a	18	1/2 of the yolk is grown over	—	63	44	
13b	24	3/4 of the yolk is grown over	70	18	16	
13c	27	Prior to closing of the blastopore	60	50	45	Malformations are of various kinds
15a	35	Closing of the blastopore	54	38	40	
15b	48	Formation of the embryo	55	27	17	
16	60	Formation of the embryo finished	10	7	0	
16a	82	Appearance of blood circulation	0	0	0	
17a	105	Hatching	10	6	0	

It is evident from the data of Table 2 that periods of different sensitivity represented by the figure may be detected only in the case of temperature of 6°C. A general decrease of sensitivity in the course of embryonic development is noticeable when 0° and 3°C are applied. This may be obviously explained by the fact that application of these temperatures represents such a strong treatment that one fails to discover periods of different sensitivity.

The following conclusion is thus reached on the basis of the above data. Different stage of embryonic development in bream react differently at one and the same harmful influence; the following periods of different sensitivity may be distinguished:

1st period—highest sensitivity, from fertilization up to the large-celled morula.

2nd period—low sensitivity, from the large-celled morula to the beginning of embryo formation.

3rd period—increased sensitivity, from the beginning of the embryo formation up to the appearance of the lens.

4th period—low sensitivity, from the appearance of the lens up to hatching.

When these periods are compared to those found in embryonic development of perch under application of high temperature ([4]) full agreement is found. The present investigation failed to discover the period of increased sensitivity at the beginning of gastrulation which was established for eggs of perch treated with high temperature([3]). This should be obviously attributed to insufficient number of experiments relating to the beginning of gastrulation.

It is necessary to note that the influence of high and low temperatures is manifested in an identical manner at identical stages; this justifies the assumption that the treatment applied is non-specific.

Application of low temperature for slowing down development of bream eggs with purely practical aims may be recommended only at later stage of development when the embryo is already formed, which takes place from 50 to 60 hours following fertilization when the latter proceeds at a temperature of 16—18°C.

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PALEOZOOLOGY

A NEW FORM OF A BIRD FROM THE PLIOCENE OF ODESSA

By V. I. ZUBAREVA

(Communicated by A. A. Borissiak, Member of the Academy, 27.III.1939)

Excavations carried out by the Odessa Paleontological Expedition of the Academy of Sciences of the Ukrainian SSR in the Odessa „catacombs" yielded a rich fauna, referable to the Middle Pliocene. It comprises a great number of remains of camel, large fox, steppe-fox, hyaena, bear, hamsters, field-mouse, beaver, piping hare, hare, hedgehog, machaerodus, mastodon and other mammals; of birds there are remains of ostriches (plentiful), a bird coming close to the marabou, of *Gallinae*, *Passers*, *Cervidae* etc.

Among bones of birds were discovered two tibiotarsal bones of striking form and robustness. These bones are so remarkable that some specialists are inclined to regard them as pathological. There is a right and left tibiotarsus of two specimens—№ 6617 (dex.) which belonged to a diminutive and № 6618 (sin.) to a rather large-sized individual. The two bones are identical as to the type of structure. Unfortunately, both of them are lacking the proximal end, beginning from the level somewhat above foramen nutritium.

The powerful distal epiphysis passes into a vaguely pronounced neck, wherefrom a diaphysis of striking form arises. Strongly compressed in the medio-lateral direction, it exhibits convexities on the distal and particularly on the proximal side. The condyles of the distal epiphysis are well developed and relatively low, condylus tibialis internus being more strongly developed.

The neck is somewhat flattened in the dorso-plantar direction, its dorso-medial end being reduced to the condition of a sharp edge. A comparatively narrow arch of ligamentum transversum ossificatum locks a narrow canalis musculus extensor digitorum communis.

A faintly discernible groove running from the canal rises obliquely upwards and outwards, passing beyond the proximal border of the lateral side. Parallel to it, starting from the neck, extends a sharp groove, evidently from the musculus peroneus, passing over to the lateral side. On the plantar side from the neck rises obliquely upwards and outwards a deep and wide groove, likewise passing to the lateral side. The grooves having transgressed on the lateral side gradually disappear.

On the lateral surface, between the proximal and distal grooves a sharp swelling is observable, becoming obliterated at the level where the grooves disappear. It is particularly well-marked on the tibiotarsus № 6617. Above this dilation there appears on the lateral surface a straight and rough line from the former articulation with the fibula at this place. Above the fora-

men nutritium and within a smaller distance from it to the plantar surface is a perceptible depression, apparently corresponding to the place of attachment of musculus flexor digitorum perforans.

The medial wall of tibiotarsus is a smooth trough originating somewhat above the neck, formed partly because of the caving in of the medial wall proper, however, mainly on account of the expansion of the dorsal and plantar walls. On the tibiotarsus № 6617 this trough seems to be divided into an upper and lower region by a tuberosity present there. The bone has a

Tibiotarsus № 6617 *Gryzaja odessana* gen. et sp. nov. from the lateral, dorsal, medial and plantar sides (from left to right).

large canal, bean-shaped on section, strongly elongated in the dorso-plantar direction. Owing to the expansion of the walls, there is no narrowing of the canal whatever.

No tibiotarsus of such a form was observed by us either among the living or the hitherto known species of fossil birds. Complete identity of structure of the two bones belonging to different individuals gives every reason to believe this structure to be normal, and not pathological. The surface of the bone is even and smooth, with no rugosities whatsoever, which usually appear as a characteristic result of a pathological process.

According to the shape of the bones, their strong flattening in the medial-lateral direction, they may be assumed to belong to a bird from the group of swimming birds.

As to the evolution of such form, the suggestion may be ventured that in one of the groups of swimming birds which gave rise to the recent order *Pygopodes*, the development of tibiotarsus was proceeding toward the acquisition on the dorsal side of a highly durable, yet at the same time very light crest for the attachment of muscles, whereas in the other group of swimming birds to which the described form pertains, for the same reason, the evolution proceeded by way of expansion of the diaphysis of the bone.

This instance may serve as an illustration of one of the so-called cul-de-sacs of evolution.

The above is a conclusive evidence, leading us to believe that the described remains of a bird belong to an original extinct form, unknown among the living as well as the fossil ornithofauna, a form we shall designate as *Gryzaja odessana* gen. et sp. nov.*

* The generic name is assigned in honour of the Chief of the Odessa Paleontological Expedition of the Academy of Sciences of the Ukrainian SSR, T. G. Gryzaj, who was very efficient in the organization of excavations and recovery of material; the specific—after the name of the locality.

Dimensions of the described remains of *Gryzaja odessana* gen. et sp. nov. are the following:

	Tibiotarsus	
	№ 6617	№ 6618
Medial-lateral measurement of the distal epiphysis	14 mm	15.8 mm
Dorso-plantar » » » » »	13.6 »	14 »
Height of condyles of the epiphysis	10.5 »	11 »
Medial-lateral measurements of the neck	11 »	11.4 »
Dorso-plantar » » » »	7.3 »	8.1 »
Width of the arch	8.4 »	8.2 »
Medial-lateral measurements of the stoutest part of the diaphysis	12.5 »	10.5 »
Dorso-plantar measurement ibid.	17 »	15.9 »
Maximum dorso-plantar measurement of the diaphysis	—	22.4 »
Medial-lateral measurement of the diaphysis ibid.	—	10.5 »

Continued paleontological excavations in Odessa are expected to supplement the amount of remains of this original bird so as to enable us to give a more complete description.

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PALEOZOOLOGY

CONTRIBUTION TO THE HISTORY OF THE FAUNA OF THE USSR

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The roots of development of the recent fauna of land vertebrates are traceable as far back as the Upper Tertiary. Among the Hipparion fauna in the south of the USSR existed already numerous genera of the contemporary fauna (*Castor*, *Hystrix*, *Spalax*, *Lepus*, *Ochotona*, *Erinaceus*, *Vulpes*, *Ursus*, *Hyaena*, *Bubo*, *Nyctala*, *Pelobates* and others), alongside of many now extinct ones (*Hipparion*, *Aceratherium*, *Mastodon*, *Dinotherium*, *Achtiaria*, *Samotherium*, *Procervus*, *Ictitherium*, *Machaerodus* etc.), or exotic as against the modern fauna of the USSR (*Elephas*, *Rhinoceros*, *Cervulus*, *Arctonyx*, *Macacus*, *Struthic* and others).

The Hipparion fauna was part of the fauna extending into subtropical and partly periboreal latitudes, encircling during the Upper Tertiary, besides Africa, Southern Europe and Asia, a considerable part of Eastern Europe and North-Eastern Asia. During the Quaternary, connected with the continental glaciation, the development of the fauna in the northwest of Europe and Asia took a course essentially different from that in Africa and South-Eastern Asia.

In the deposits of the Middle and Upper Pliocene in the south of the European part of the USSR occur among the rather impoverished Hipparion fauna: monkeys, muntjac (*Cervulus*), arctonyx, ostrich, a bird close to the Indian marabou and other forms, having their nearest relatives among the contemporaneous fauna of Africa and South-Eastern Asia, but non-existent even in the Early Quaternary deposits in the south of the USSR. In the deposits of the earliest Quaternary in the south of the European part of the USSR are found specimens of *Elasmotherium*, mastodons (*Mastodon arvernensis*) and an elephant (*Elephas meridionalis*), showing some affinities with the living Indian elephant and characters of rapid evolution toward the mammoth group of elephants.

The mammoth is the most common fossil in the Quaternary deposits of the USSR, and the question concerning the mammoth and the entire faunal complex associated with it, called the mammoth fauna, is one of the principal questions of the paleontology of the Quaternary. The mammoth fauna directly ensues from the Hipparion fauna, which has lost most of its typical representatives.

On the confines of the Tertiary and Quaternary during the Late Pliocene, in the south of the European part of the USSR took place the ultimate extinction of such representatives of the Hipparion fauna as hipparion,

monkeys, machairodont, ostrich etc. Remains of these animals, e. i. hipparion and monkeys, do not reach above the Kuchurgan sand and gravel deposits whose origin is linked on to the Pliocene glaciation of the Carpathians. The existence of a great continental, so-called Günz glaciation in the USSR and adjoining regions of Middle and Western Europe during the Pliocene assumed by some geologists has not been borne out by paleontological evidence. The rich fauna unearthed in the Odessa „catacombs", no older in age than the Middle Pliocene, comprises a great number of *Ungulata*, *Carnivora* and rodents, but among these animals there are neither any representatives, nor nearest ancestry of arctic animals, for instance, reindeer, arctic fox, lemmings etc. Quite the contrary, the main background of this fauna is made up of animals of the peribореal, and to some extent, subtropical type, like camels, mastodons, hyaena, arctonyx, ostriches etc.

The end of the Pliocene was marked by a notable change of landscape and climatic conditions, due to the changed position of the arctic region which was drawing closer to the north-western outskirts of Europe. This change is well perceived in the transition from the Hipparion to the Mammoth fauna. As early as in the Pliocene among the elephants of the group *Elephas meridionalis* are traced individuals with lamellae on the molars increasing in number, the enamel walls of the plates respectively growing thinner.

This process of formation of elephants of the mammoth group, the latter being connected with the boreal landscape-geographical region, took an irregular course of development. The most ancient mammoth was found in the Pliocene of France(1) and is liable to occur in the Pliocene and Lower Quaternary deposits of other parts of Northern and Western Europe.

In sand and gravel deposits of the early Pleistocene (early Quaternary) between the cities of Nogaisk and Berdyansk, together with *Elasmotherium sibiricum*, *Cervus giganteus*, *Mustela nivalis*, *Ochotona pusilla* etc., have been recovered elephants having on the grinding surface of the molars 5—6 plates in a 10 cm line.

In the Pleistocene sand and gravel deposits of Tiraspol, among elephants identical with those of Berdyansk mentioned above, occur also individuals displaying on the grinding surface of the molars 7.5—8 plates in a 10 cm line (the total number of plates on the grinding surface reaching 16). Since mammoths with molars having 6.1—7.5 plates in a 10 cm line of the grinding surface are not infrequently met in the loess of the Middle and Upper Quaternary in the Ukrainian SSR (Belenkoye, Novogeorgyevsk, Lubny, Bobrik), as well as in the Late Quaternary deposits in the north of the Asiatic part of the USSR (e. g. the known Berezovsk mammoth), it is impossible to sharply discriminate between elephants of the Early and Middle Quaternary (the so-called *Elephas trogonthorii*) and the mammoth.

By designating the mammoth as *Elephas primigenius primigenius* Blum. (mainly Siberian mammoths) and the earlier form as *Elephas primigenius trogonthorii* Pohl. (for the most part, European mammoths of the Early and Middle Quaternary), we are thus emphasizing their common specific position and placing within natural limits the so-called aberrant specimens occurring in the deposits of the Early, Middle and Upper Quaternary.

The actual material available from the southern part of the USSR warrants the statement that the mammoth and mammoth fauna occurred there by way of evolution of forms which in the main body had already been part of the late Hipparion fauna. As the immediate ancestor of the mammoth proper should be regarded the southern elephant *Elephas meridionalis* Nesti.

The well-known fauna of the Tiraspol gravel (from the Kolkotov quarries), as already mentioned, furnishes, alongside of elephants close to *Elephas meridionalis*, forms indiscernible from the mammoth. By referring

the Tiraspol fauna to the Middle Pleistocene (the so-called Mindel) we are in fact determining also the beginning of formation of the fauna named by us mammoth fauna. The latter comprised *Elephas primigenius*, *Bison priscus*, *Equus equus*, *Cervus elaphus*, *Alces*, *Capreolus*, *Felis leo*, *Ursus spelaeus*, *Vulpes vulpes*, *Hyaena crocuta*, *Lepus europaeus*, *Ochotona pusilla*, *Castor fiber*, *Spalax leucodon*, *Cricetulus migratorius*, *Citellus pygmaeus*, *Lagurus lagurus*, *Erinaceus* and others. By growing poorer and suffering slight modification, this fauna gave rise to successive faunas including the recent one (*Muridae*).

The above statements regarding the fauna, coupled with the fact that nearly the entire contemporary European part of the USSR presented in the Lower Quaternary dry land, makes us assume that the range of the mammoth and the groups of animals (mammoth fauna) associated with it extended into the northernmost parts of the continent of today.

It is believed that for want of actual facts it is impossible to prove that during the preglacial, i. e. the Early Quaternary period, the mammoth had spread throughout the entire European part of the USSR. However, this view conflicts with the finds of sub-moraine mammoth remains in intermoraine deposits (if there be several boulder horizons) and in sub-moraine fluvioglacial deposits in the middle and northern part of the USSR, as well as in Scandinavia (sub-moraine), testifying that by the time the Ice Age set in, the mammoth had already existed. The extinction of the mammoth went along with the glaciation.

The analysis of the rich „mixed" fauna found in the glacial till and the lower part of the supra-moraine loess in Novgorod-Seversk and Mezino of the Chernigov province has shown that the arctic group of animals [arctic fox, reindeer, musk-ox (*Ovibos moschatus*), collared lemmings, snow partridge, arctic owl] which appeared simultaneously with glaciation to coexist with the native steppe fauna [jeroba, bobak (polish marmot), susliks (*Citellus rufescens*), small piping hare, yellow and grey steppe lemmings (*Lemmus lemmus*), diminutive hamster etc.], whereby the indigenous forms partly died out, partly adapted themselves to the periglacial conditions of life (susliks, bobak)(2). A similar phenomenon occurred in the subsequent epoch (i. e. in the Late Quaternary) in Noth-Eastern Siberia, where the arctic fauna which we have named dicrostonyx, outsted the native fauna of the steppe type, and hence such animals as the suslik and bobak came to live in the tundra, having adapted themselves to the local conditions of life.

In the light of the foregoing it is clear why under the conditions of the progressive, stationary and regressive phases of glaciation we find remains of mammoths in association with both arctic and steppe forms.

During the Middle Quaternary the landscape-geographical zones of the European part of the USSR were shifted southward and eastward, according to the latitudes of that time following the south-west to north-east course. In connexion therewith, the forest and forest-steppe fauna extended its range into the Crimea and Caucasus, and during seasonal migrations representatives of the dicrostonyx fauna (for example, the reindeer and arctic fox in the Crimea)(3) reached the very same boundaries.

Owing to the thick beds of loess-like and arenaceous deposits which had formed chiefly at the expense of the fluvioglacial material, there was a sharp change in the ecological conditions in the south of the USSR, as compared to the preceding epochs. The mammoth and rhinoceros died out completely within that area, and the place of the mammoth fauna was taken by that of bison-deer which has existed up to the beginning of our era. With the extinction of bison and deer in the European part of the USSR the dominant position, both in regard to the number of species and specimens, has

been taken up by rodents and other diminutive animals—the recent fauna *Muridae* has arisen.

Thus, the development of the vertebrate fauna furnishes clear evidence of the great Quaternary wave of cold (Middle Quaternary) which led to the extinction of a considerable number of representatives of the mammoth fauna in the glacial and periglacial regions. In regions not subjected to the direct or indirect action of the great Quaternary continental glaciation of Europe and south-western part of Asia (extending to Taimyr) the mammoth fauna including the mammoths and rhinoceroses was still existent in the Upper Quaternary.

The existence of the mammoth in the north-east of Asia in association with warmth-loving flora, antecedent to the recent refrigeration, is confirmed by the occurrence in the Yakutsk ASSR of the grey American nut (*Juglans cinerea* L.) alongside of mammoth remains([4]).

The retreat of the dicrostonyx fauna in the Late Quaternary epoch to the contemporaneous northern regions occurred not at once. As early as in the Neolithic age (i. e. about 4000—5000 years before our time) there lived in the Chernigov province of the Ukrainian SSR the arctic fox, and in the Kiev province—the reindeer; relicts of the common hare *Lepus timidus* and snow partridge (*Lagopus lagopus*) are existent in the Chernigov province even now.

The retreat of some representatives of the dicrostonyx fauna northward, simultaneously with the recession of the secular frozen grounds in the north-east of the European part of the USSR is now still going on.

As indicated above, the formation and development of the arctic fauna in the European part of the USSR and in the north-east of the Asiatic part of the USSR are not coeval. The extinction and the entombement in the frozen soil of remains of such animals as the mammoth, rhinoceros, saiga-antelope, tiger in the north-east of Asia (Novosibirsk Islands) ([5]) occurred in the Late Quaternary.

The presence in the Middle Quaternary, and possibly, the Late Quaternary as well, of the ostrich and antelope of the African type([7]) in Transbaikalia([6]) and Mongolia, is taken by us as a proof of the existence of holarctic conditions at the time when continental glaciation prevailed in Europe.

The occurrence in the Quaternary deposits of Alaska of saiga-antelope, yak([8]) and camel([9]) bears witness to the continental relationship between the fauna of the north-east of Asia and of Alaska, which lasted until the Late Quaternary refrigeration in this part of Asia and America.

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